

PATENT ABSTRACTS OF JAPAN

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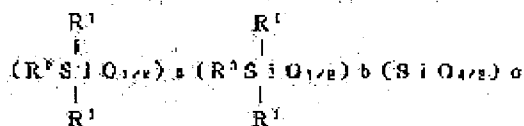
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(54) CURABLE RESIN COMPOSITION

(57)Abstract:

PURPOSE: To obtain the subject composition having excellent flowability and giving a cured resin having excellent flexibility and adhesivity by compounding a curable resin with a specific amount of an organopolysiloxane having an epoxy-containing organic group.

CONSTITUTION: The objective composition can be produced by uniformly mixing (A) 100 pts.wt. of a curable material (preferably epoxy resin, phenolic resin, etc.,) with (B) 0.1-500 pts.wt. of a compound of the formula [R1 is a univalent hydrocarbon group excluding alkenyl; R2 is H or a univalent hydrocarbon group excluding alkenyl; R3 is an epoxy-containing organic group or an alkoxysilylalkyl; (a) is ≥0; (b) and (c) are positive numbers; a/c is 0-4; b/c is 0.05-4; (a+b)/c is 0.2-4] using a single screw or twin-screw continuous mixer, etc.



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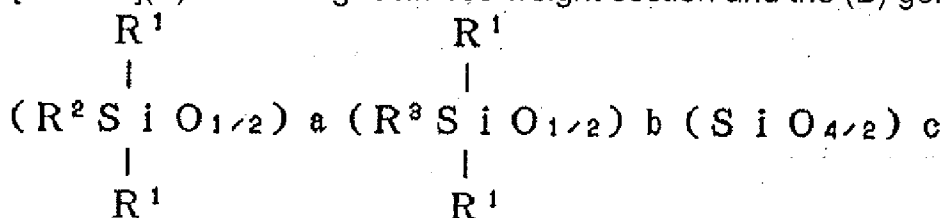
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CLAIMS

[Claim(s)]

[Claim 1](A) Hardening resin 100 weight section and the (B) general formula : [Formula 1]



{R¹ is a monovalent hydrocarbon group except an alkenyl group among a formula, and R² is the monovalent hydrocarbon group or hydrogen atom except an alkenyl group, R³ is an epoxy group content organic group or an alkoxy silyl alkyl group, and a is 0 or a positive number, b is a positive number, c is a positive number, a/c is the number of 0-4, and b/c is the number of 0.05-4, and (a+b)/c is the number of 0.2-4. The hardenability resin composition which consists of 0.1 to organopolysiloxane 500 weight section which is expressed with}, and which has an epoxy group content organic group.

[Claim 2](A) The hardenability resin composition according to claim 1 which is hardening resin chosen from a group which hardening resin of an ingredient becomes from an epoxy resin, phenol resin, imide resin, and silicone resin.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Industrial Application] This invention is excellent in mobility before hardening in detail about a hardenability resin composition, and after hardening is related with the hardenability resin composition used as hardening resin excellent in flexibility and an adhesive property.

[0002]

[Description of the Prior Art] Since the hardening resin produced by hardening this is excellent in mechanical properties, such as electrical properties, such as dielectric characteristics, volume resistivity, and electrical breakdown strength, or flexural strength, compressive strength, and impact strength, the hardenability resin composition is used for the electrical and electric equipment and sealing resin for electronic devices, adhesives, resin for FRP, etc.

[0003] Generally, a hardenability resin composition has the upright hardening resin produced by hardening this, and Since [lacking in flexibility and] the rate of cure shrinkage at the time of hardening is large, If this is used as the electrical and electric equipment and sealing resin for electronic devices, in order to add big stress to the electrical and electric equipment and an electronic device, There was a problem of producing a crack in hardening resin itself, or destroying the electrical and electric equipment and an electronic device, and producing a crevice between the electrical and electric equipment and an electronic device, and hardening resin since the adhesive property over the electrical and electric equipment and electronic device of hardening resin is low. The coefficient of thermal expansion of the hardening resin produced by hardening this a hardenability resin composition to the coefficient of thermal expansion of the electrical and electric equipment and an electronic device Since it is large, If the resin seal electrical and electric equipment and an electronic device receive a thermo cycle, in order to add big stress to the electrical and electric equipment and an electronic device, There was a problem of producing a crack in hardening resin itself, or destroying the

electrical and electric equipment and an electronic device, and producing a crevice between the electrical and electric equipment and an electronic device, and hardening resin since the adhesive property over the electrical and electric equipment and electronic device of hardening resin is low. When water invaded into the crevice between the crack or hardening resin produced by the cure shrinkage of a hardenability resin composition, or thermal expansion contraction, and the electrical and electric equipment and an electronic device, there was a problem of promoting degradation of the resin seal electrical and electric equipment and an electronic device.

[0004] Thus, in the hardenability resin composition, in order to low-stress-ize hardening resin, the flexibility of hardening resin is raised, and the hardenability resin composition of the versatility which makes the adhesive property over the electrical and electric equipment and electronic device of hardening resin come to improve is proposed. For example, an epoxy resin, a methylphenyl polysiloxane, the organopolysiloxane that consists of bifunctional siloxane units (D unit) which have an epoxy group, and trifunctional siloxane units (T unit), The hardenability resin composition which consists of an inorganic filler and a catalyst for hardening (refer to JP,56-145942,A), . Have an epoxy resin, an epoxy group or an amino group and a hydroxyl group, or at least one basis that can be hydrolyzed, respectively. The organopolysiloxane which consists of monofunctional siloxane units (M unit) or trifunctional siloxane units (T unit), and bifunctional siloxane units (D unit), The hardenability resin composition which consists of a hardening agent and an inorganic filler (refer to JP,56-136816,A), An epoxy resin, phenol resin, the hardenability resin composition that becomes a chain side chain from the dimethylpolysiloxane which has an epoxy group and a polyoxyalkylene group (refer to JP,60-58425,A), And the hardenability resin composition (refer to JP,64-51467,A) which consists of hardening resin and silicone rubber spherical pulverized coal is proposed.

[0005] However, the hardenability resin composition proposed by JP,56-145942,A and JP,56-136816,A, Since the organopolysiloxane which makes bifunctional siloxane units (D unit) indispensable is used, There was a fault that the glass transition point (T_g) of the obtained hardening resin fell, and it was remarkable especially when using Loki Sun for the organopolysiloxane which consists of $2\text{SiO}_2/a_2$ unit (D unit) which has an epoxy group (CH_3).

The hardenability resin composition proposed by JP,60-58425,A, In order to use the dimethylpolysiloxane which has an epoxy group and a polyoxyalkylene group for a chain side chain, there was a fault that the obtained hardening resin had large absorptivity and it was not suitable as the electrical and electric equipment and sealing resin for electronic devices. The hardenability resin composition proposed by JP,64-51467,A, Although the flexibility of the hardening resin produced by hardening this was excellent, since the mobility of a hardenability resin composition was bad, there was a fault that the workability which carries out the resin

seal of the electrical and electric equipment and the electronic device was bad, and this hardening resin had a still worse adhesive property over the electrical and electric equipment and an electronic device.

[0006]

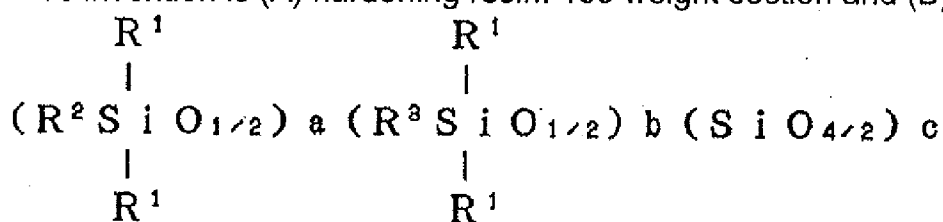
[Problem(s) to be Solved by the Invention] This invention persons reached this invention, as a result of trying hard wholeheartedly, in order to solve the above-mentioned problem.

[0007] That is, the purpose of this invention is excellent in mobility before hardening, and there is after hardening in providing the hardenability resin composition used as hardening resin excellent in flexibility and an adhesive property.

[0008]

[The means for solving a problem, and its operation]

This invention is (A) hardening resin. 100 weight section and (B) general formula : [Formula 2]



{R¹ is a monovalent hydrocarbon group except an alkenyl group among a formula, and R² is the monovalent hydrocarbon group or hydrogen atom except an alkenyl group, R³ is an epoxy group content organic group or an alkoxy silyl alkyl group, and a is 0 or a positive number, b is a positive number, c is a positive number, a/c is the number of 0-4, and b/c is the number of 0.05-4, and (a+b)/c is the number of 0.2-4. It is related with the hardenability resin composition which consists of 0.1 to organopolysiloxane 500 weight section which is expressed with}, and which has an epoxy group content organic group.

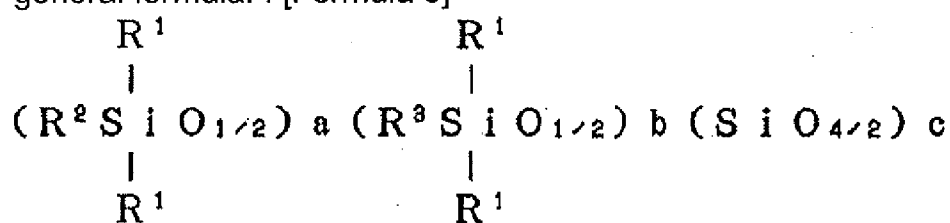
[0009] Hereafter, the hardenability resin composition of this invention is explained in detail.

[0010] (A) An ingredient is hardening resin which is a substrate of this invention. (A) As an ingredient, publicly known hardening resin can be used conventionally. (A) As an ingredient specifically, Phenol resin, formaldehyde resins, xylene resin, xylene-formaldehyde resin, ketone formaldehyde resins, furan resin, urea resin, imide resin, melamine resin, an alkyl resin, unsaturated polyester resin, aniline resin, sulfone amide resin, Silicone resin, epoxy resins, and these copolymer resin are illustrated, and it can be used combining one sort of the hardening resin of the above-mentioned illustration, or two sorts or more. It is preferred that it is hardening resin chosen from the group which consists of an epoxy resin, phenol resin, imide resin, and silicone resin especially as a (A) ingredient. The hardening resin etc. in which the curing means hardens especially the (A) ingredient with the hardening resin hardened with high energy beams which it is not limited but are hardened with heat, such as hardening resin,

ultraviolet rays, or radiation, and humidity are illustrated. At a room temperature, the (A) ingredient may be liquefied or may be hardening resin of which state of a solid state.

[0011](A) A hardening agent, a hardening accelerator, a bulking agent, a photosensitizer, higher-fatty-acid metal salt, an ester system wax, a plasticizer, etc. can be blended with an ingredient as ingredients other than the hardening resin of the above-mentioned illustration. As a hardening agent, specifically Organic acid, such as carboxylic acid and sulfonic acid, and the anhydride of those, an organic hydroxy compound, a silanol group, The amino compound of the organic silicon compound and the first class which have bases, such as an alkoxy group or a halogeno group, or the second class can be illustrated, and it can be used combining these kinds or two sorts or more. Specifically as a hardening accelerator, organophosphorus compounds, such as organic metallic compounds, such as the third class amine compound, aluminum, and a zirconium, and phosphine, a different ring type amine compound, a boron complex compound, organic ammonium salt, organic sulfonium salt, organic peroxide, etc. are illustrated. As a bulking agent, specifically, Glass fiber, asbestos, an alumina fiber, alumina, and silica. Fibrous fillers, such as the ceramic fiber used as an ingredient, a boron fiber, a zirconia fiber, silicon carbide fiber, a metal fiber, polyester fiber, an aramid fiber, a nylon fiber, a phenol fiber, and natural animals-and-plants textiles, fused silica, precipitation silica, fumed silica, pyrogenic silica, a zinc oxide, Calcination clay, carbon black, a glass bead, alumina, talc, calcium carbonate, clay, aluminium hydroxide, barium sulfate, a titanium dioxide, aluminium nitride, silicon carbide, magnesium oxide, beryllium oxide, kaolin, mica, Particulate matter-like bulking agents and these mixtures, such as zirconia, are illustrated.

[0012](B) An ingredient is an ingredient for raising the flexibility of the hardening resin produced by hardening the hardenability resin composition of this invention, and raising the adhesive property over the electrical and electric equipment and an electronic device, and is a general formula. : [Formula 3]



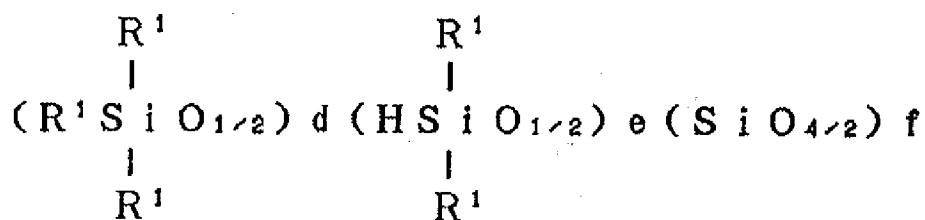
It is organopolysiloxane come out of and expressed. R^1 is a monovalent hydrocarbon group except an alkenyl group among an upper type, and specifically, A chloromethyl group, 3, 3, Aralkyl groups, such as aryl group; benzyls, such as alkyl group; phenyl groups, such as a methyl group, an ethyl group, a propyl group, a butyl group, a pentyl group, a hexyl group, and an octyl group, a tolyl group, and a xylyl group, and a phenethyl group; Substituted alkyl groups, such as 3-trifluoropropyl group, etc. are illustrated. R^2 is the monovalent hydrocarbon

group or hydrogen atom except an alkenyl group, and as a monovalent hydrocarbon group except an alkenyl group specifically, A chloromethyl group, 3, 3, Aralkyl groups, such as aryl group; benzyls, such as alkyl group; phenyl groups, such as a methyl group, an ethyl group, a propyl group, a butyl group, a pentyl group, a hexyl group, and an octyl group, a tolyl group, and a xylyl group, and a phenethyl group; Substituted alkyl groups, such as 3-trifluoropropyl group, etc. are illustrated. R^3 is an epoxy group content organic group or an alkoxy silyl alkyl group, and as an epoxy group content organic group specifically, A glycidoxo ethyl group, a glycidoxo propyl group, A glycidoxobutyl group, a 3,4-epoxycyclohexyl ethyl group, a 3,4-epoxycyclohexyl propyl group, a 3,4-epoxy NORUBORUNE nil ethyl group, a 2-(3,4-epoxy-3-methylcyclohexyl)-2-methylethyl group, etc. are illustrated, As an alkoxy silyl alkyl group, specifically, A trimethoxysilyl ethyl group, a triethoxy silyl ethyl group, a methyl dimethoxy silyl ethyl group, a trimethoxysilylpropyl group, a triethoxy silyl propyl group, a trimethoxysilyl butyl group, a methyl dimethoxy silyl propyl group, etc. are illustrated.

[0013]a is 0 or the positive number which shows the number of the monofunctional siloxane units (M unit) which do not have an epoxy group content organic group or an alkoxy silyl alkyl group among an upper type, b is a positive number which shows the number of the monofunctional siloxane units (M unit) which have an epoxy group content organic group or an alkoxy silyl alkyl group, c is a positive number which shows the number of tetrafunctional siloxane units (Q unit), each ratio and a/c are the number of the ranges of 0-4, and b/c is the number of the ranges of 0.05-4, and (a+b)/c is the number of the ranges of 0.2-4. Since monofunctional siloxane units (M unit) cannot have this [good] for four pieces to one tetrafunctional siloxane units (Q unit) and compatibility is excellent in the (B) ingredient to the (A) ingredient in this invention, It is because the monofunctional siloxane units (M unit) which have an epoxy group content organic group or an alkoxy silyl alkyl group need to be at least 0.05 piece to one tetrafunctional siloxane units (Q unit). (B) Although organopolysiloxane of an ingredient is liquefied at a room temperature, or it is a solid state and the molecular weight in particular is not limited, since compatibility with the (A) ingredient is good, it is preferred that it is the range of 500-500,000.

[0014]In this invention, organopolysiloxane of the (B) ingredient, Since it has an epoxy group content organic group or an alkoxy silyl alkyl group, (A) Since the flexibility of the hardening resin obtained by being incorporated into the resin matrix of an ingredient improves and the (B) ingredient consists of a SiO_2 unit (Q unit), the glass transition point (T_g) of the obtained hardening resin does not get worse, but has the advantage of excelling in heat resistance and fire retardancy.

[0015]Organopolysiloxane of the above-mentioned (B) ingredient is the bottom of existence of the catalyst for a hydrosilylation reaction, and a general formula, for example. : [Formula 4]



{R¹ is a monovalent hydrocarbon group except an alkenyl group among a formula, d is 0 or a positive number, e is a positive number, f is a positive number, d/f is the number of 0-4, and e/f is the number of 0.05-4, and (d+e)/f is the number of 0.2-4. It can obtain by carrying out the addition reaction of the alkoxy silylalkene of the organic compound which has organopolysiloxane and the epoxy group which have a silicon atom absorbed water matter atom, and aliphatic unsaturated bonds which are expressed with}, and an arbitrary dose.

[0016](B) The loadings of an ingredient are 0.1 - 500 weight section to (A) ingredient 100 weight section.

It is 0.5 to 100 weight section preferably.

This is because there is no effect that the loadings of the (B) ingredient are less than 0.1 weight sections to (A) ingredient 100 weight section in low stress-ization of hardening resin. It is for the mechanical strength of hardening resin to fall remarkably, if 500 weight sections are exceeded.

[0017]The hardenability resin composition of this invention is obtained by mixing the (A) ingredient and the (B) ingredient uniformly. (A) The method in particular of mixing an ingredient and the (B) ingredient is not limited, but specifically, (A) After blending and carrying out the premix of the (B) ingredient at the time of the method of blending the (B) ingredient with an ingredient directly, and (A) ingredient preparation, the method of blending additive agents, such as a bulking agent, with the (A) ingredient and the method of blending with the (B) ingredient various kinds of additive agents blended with the (A) ingredient one by one are illustrated. (A) Although there is no limitation and a mixed device is chosen by the gestalt of versatility [ingredient / the (A) ingredient or / (B)], such as a liquid, a solid state, and powder, especially as a mixed device which mixes an ingredient and the (B) ingredient, Specifically, the continuous mixer of one axis or two axes, 2 rolls, a loss mixer, and a kneader mixer are illustrated.

[0018]Since the hardenability resin composition of this invention is excellent in mobility before hardening, it can be used by methods, such as a transfer mold, injection molding, potting, casting, powder coating, dip coating, and dropping. Since after hardening becomes hardening resin excellent in flexibility and an adhesive property, the hardenability resin composition of this invention can be used for the hardenability resin composition for closing the electrical and electric equipment and an electronic device, adhesives, etc.

[0019]

[Example] Hereafter, an example explains this invention. The value of viscosity is a value measured in 25 ** among an example, and the various characteristics of a hardenability resin composition and hardening resin were measured by the method shown below.

[0020] O Spiral flow : it measured by the method according to an EMMI standard.

O Molding shrinkage : It measured by the method according to JIS-K-6911.

O Coefficient of thermal expansion : The hardening resin fabricated to 5mmx5mmx16mm was measured using the thermal expansion meter (DL-7000 by a vacuum science-and-engineering company). The value of a coefficient of thermal expansion is a value from a room temperature to a glass transition point.

O Glass transition point (Tg) : it measured by coefficient-of-thermal-expansion measurement.

O Rate of bending flexibility : It measured by the method according to JIS-K-6911.

O Water absorption : After making into the temperature of 121 **, and 100% of humidity the hardening resin fabricated to 2 inch x0.5 inch x0.25 inch and humidifying it for 20 hours, it asked by measuring the weight increment of the hardening resin.

O Barricade : The 20-micrometer-deep slot was used and barricade length was measured.

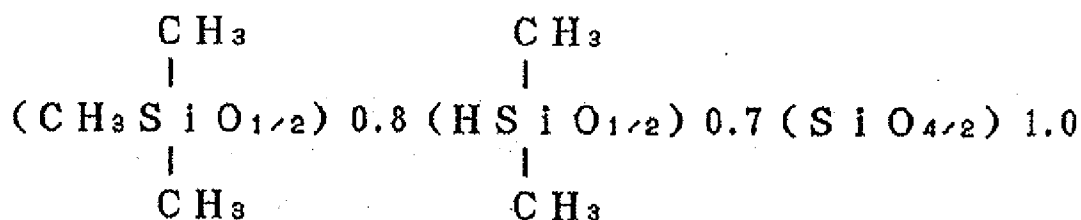
O Adhesive property : After inserting the hardenability resin composition into 42 alloy board and the copper plate and stiffening this, the adhesive property at the time of removing hardening resin was judged by O (good) and x (poor).

O Thermal shock resistance : 20 resin seal semiconductor devices (chip size 36 mm², and package 2.0 mm in thickness) are fabricated, This was performed at -196 **<-->+150 **, the heat cycle test was done in the cycle for 1 minute, the sealing resin surface was observed with the stereoscopic microscope after 150 cycles, and the case where they were ** and 11 pieces or more about the case where they are O and 6-10 pieces about the case where the number of the mold goods in which the crack went into the surface is five or less pieces was made into x.

O Solder heat resistance : 20 resin seal semiconductor devices (chip size 36 mm², and package 2.0 mm in thickness) were fabricated, this was promptly immersed in a 240 ** solder bath after 72-hour neglect under 85%RH conditions at 85 ** for 1 hour, and the sealing resin surface was observed with the stereoscopic microscope. The case where they were ** and 11 pieces or more about the case where they are O and 6-10 pieces about the case where the number of the mold goods in which the crack went into the surface is five or less pieces was made into x.

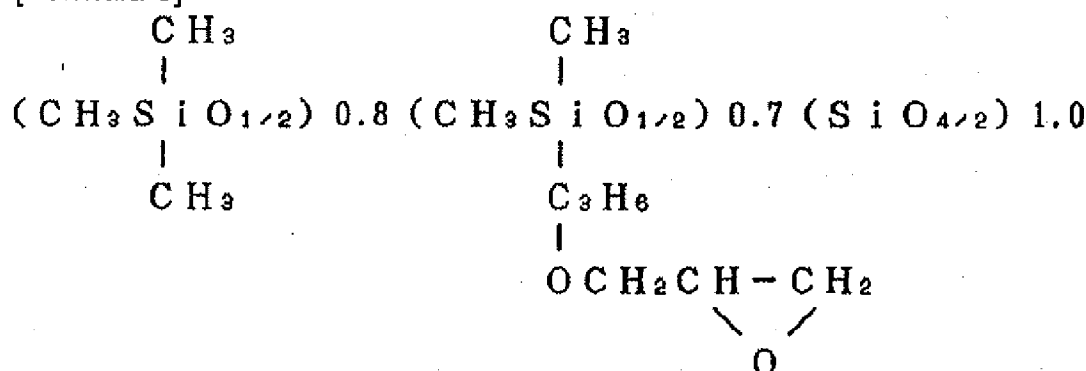
[0021]

[Reference example 1] It is a formula to agitating equipment, a flowing-back condenser tube, and a 4 mouth flask with a thermometer. : [Formula 5]



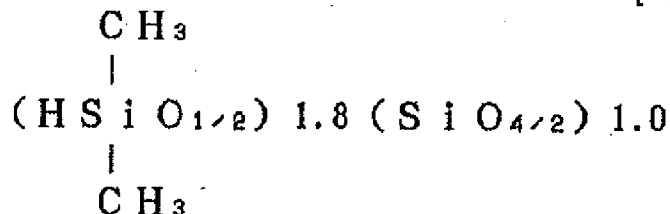
Come out and the organopolysiloxane (0.40 % of the weight of content of 105 centipoise viscosity and a silicon atom absorbed water matter atom) expressed in toluene, Allyl glycidyl ether (the number of mols of the allyl group in allyl glycidyl ether is an excessive amount to the number of mols of the silicon atom absorbed water matter atom in organopolysiloxane.) was made to react under existence of a chloroplatinic acid catalyst, and transparent brown organopolysiloxane was prepared. The viscosity of this organopolysiloxane was 520 centipoises and the weight per epoxy equivalent was 420. It was checked that this organopolysiloxane is organopolysiloxane expressed with a lower type by ^1H -nuclear-magnetic-resonance-spectrum, ^{13}C -nuclear-magnetic-resonance-spectrum, and ^{29}Si -nuclear magnetic resonance SUPEKURU.

[Formula 6]



[0022]

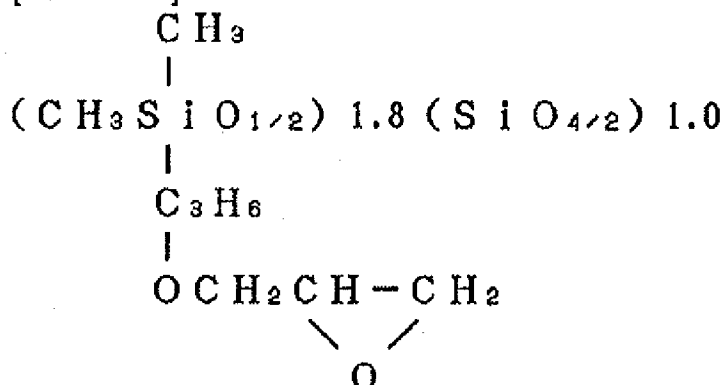
[Reference example 2] It is a formula to agitating equipment, a flowing-back condenser tube, and a 4 mouth flask with a thermometer. : [Formula 7]



Come out and the organopolysiloxane (0.92 % of the weight of content of 46 centipoise viscosity and a silicon atom absorbed water matter atom) expressed in toluene, Allyl glycidyl ether (the number of mols of the allyl group in allyl glycidyl ether is an excessive amount to the number of mols of the silicon atom absorbed water matter atom in organopolysiloxane.) was

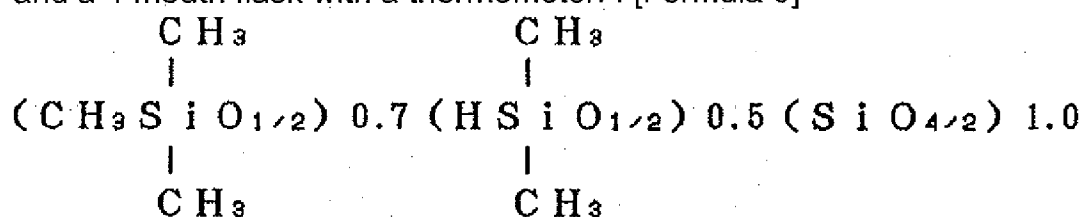
made to react under existence of a chloroplatinic acid catalyst, and transparent brown organopolysiloxane was prepared. The viscosity of this organopolysiloxane was 610 centipoises and the weight per epoxy equivalent was 370. It was checked that this organopolysiloxane is organopolysiloxane expressed with a lower type by ^1H -nuclear-magnetic-resonance-spectrum, ^{13}C -nuclear-magnetic-resonance-spectrum, and ^{29}Si -nuclear magnetic resonance SUPEKURU.

[Formula 8]



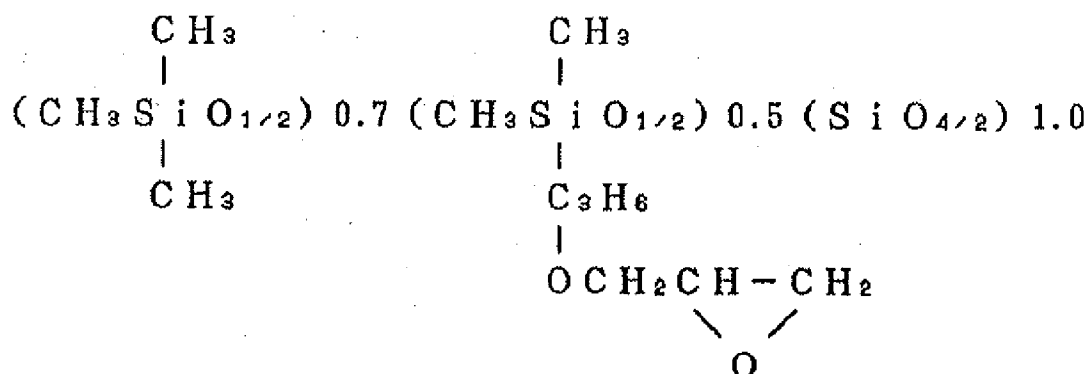
[0023]

[Reference example 3] It is a formula to agitating equipment, a flowing-back condenser tube, and a 4 mouth flask with a thermometer. : [Formula 9]



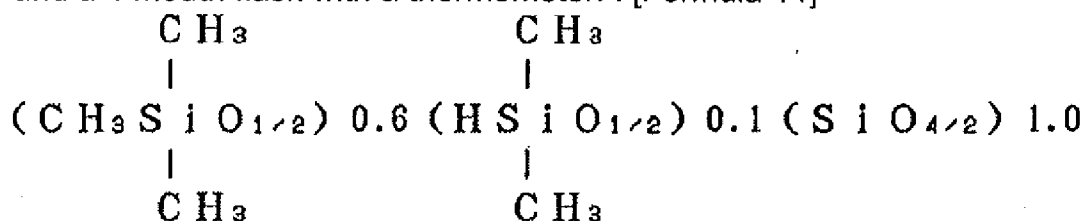
Come out and the consistency organopolysiloxane (0.33 % of the weight of content of a silicon atom absorbed water matter atom) expressed in toluene, Allyl glycidyl ether (the number of mols of the allyl group in allyl glycidyl ether is an excessive amount to the number of mols of the silicon atom absorbed water matter atom in organopolysiloxane.) was made to react under existence of a chloroplatinic acid catalyst, and organopolysiloxane of the brown shape of a translucent starch syrup was prepared. The weight per epoxy equivalent of this organopolysiloxane was 1100. It was checked that this organopolysiloxane is organopolysiloxane expressed with a lower type by ^1H -nuclear-magnetic-resonance-spectrum, ^{13}C -nuclear-magnetic-resonance-spectrum, and ^{29}Si -nuclear magnetic resonance SUPEKURU.

[Formula 10]



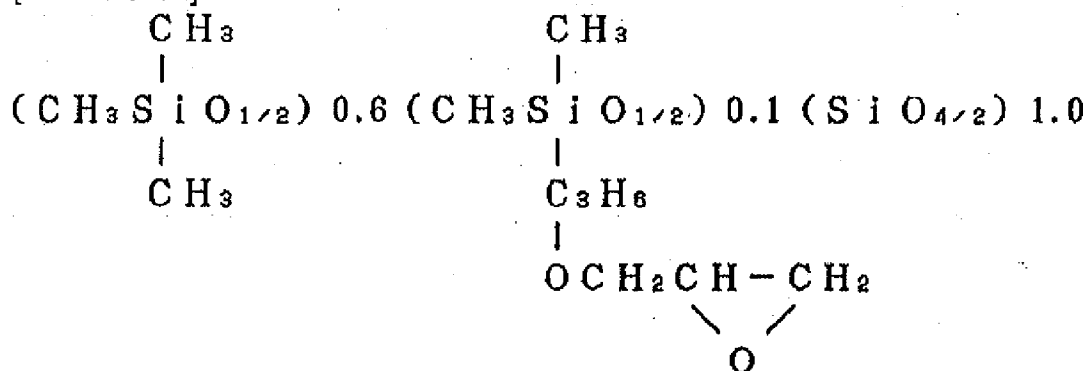
[0024]

[Reference example 4] It is a formula to agitating equipment, a flowing-back condenser tube, and a 4 mouth flask with a thermometer. : [Formula 11]



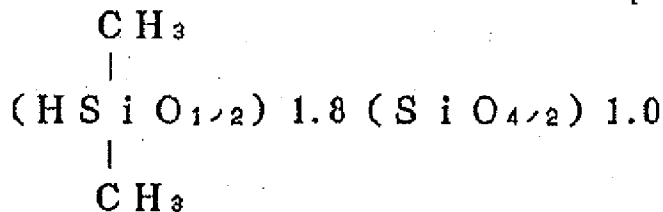
Come out and to 68% of the weight of the toluene solution of the organopolysiloxane (0.09 % of the weight of content of a silicon atom absorbed water matter atom) expressed. Allyl glycidyl ether (the number of mols of the allyl group in allyl glycidyl ether is an excessive amount to the number of mols of the silicon atom absorbed water matter atom in organopolysiloxane.) was made to react under existence of a chloroplatinic acid catalyst, and organopolysiloxane of the brown shape of a translucent starch syrup was prepared. The weight per epoxy equivalent of this organopolysiloxane was 1290. It was checked that this organopolysiloxane is organopolysiloxane expressed with a lower type by ^1H -nuclear-magnetic-resonance-spectrum, ^{13}C -nuclear-magnetic-resonance-spectrum, and ^{29}Si -nuclear magnetic resonance SUPEKURU.

[Formula 12]



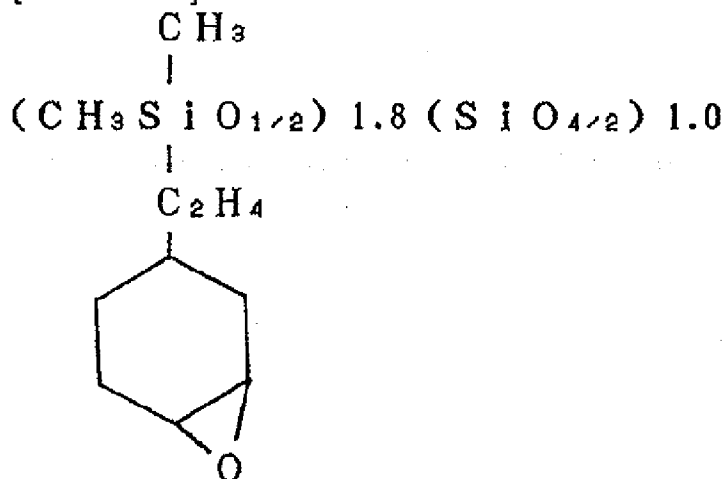
[0025]

[Reference example 5] It is a formula to agitating equipment, a flowing-back condenser tube, and a 4 mouth flask with a thermometer. : [Formula 13]



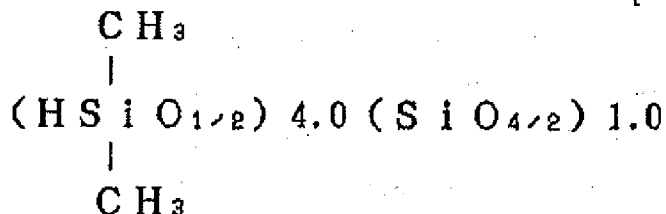
Come out and the organopolysiloxane (0.92 % of the weight of content of 46 centipoise viscosity and a silicon atom absorbed water matter atom) expressed in toluene, 1,2-epoxy-4-vinylcyclohexane (to the number of mols of the silicon atom absorbed water matter atom in organopolysiloxane) The number of mols of the vinyl group in 1,2-epoxy-4-vinylcyclohexane is an excessive amount. It was made to react under existence of a chloroplatinic acid catalyst, and transparent brown organopolysiloxane was prepared. The viscosity of this organopolysiloxane was 520 centipoises and the weight per epoxy equivalent was 230. It was checked that this organopolysiloxane is organopolysiloxane expressed with a lower type by ^1H -nuclear-magnetic-resonance-spectrum, ^{13}C -nuclear-magnetic-resonance-spectrum, and ^{29}Si -nuclear magnetic resonance SUPEKURU.

[Formula 14]



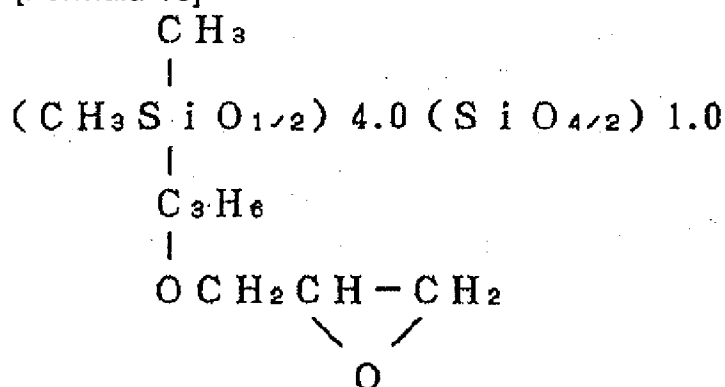
[0026]

[Reference example 6] It is a formula to agitating equipment, a flowing-back condenser tube, and a 4 mouth flask with a thermometer. : [Formula 15]



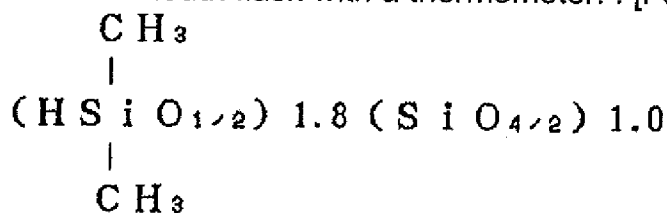
Come out and the organopolysiloxane (1.22 % of the weight of content of 190 ** of boiling points, and a silicon atom absorbed water matter atom) expressed in toluene, Allyl glycidyl ether (the number of mols of the allyl group in allyl glycidyl ether is an excessive amount to the number of mols of the silicon atom absorbed water matter atom in organopolysiloxane.) was made to react under existence of a chloroplatinic acid catalyst, and transparent brown organopolysiloxane was prepared. The viscosity of this organopolysiloxane was 54 centipoises and the weight per epoxy equivalent was 205. It was checked that this organopolysiloxane is organopolysiloxane expressed with a lower type by ^1H -nuclear-magnetic-resonance-spectrum, ^{13}C -nuclear-magnetic-resonance-spectrum, and ^{29}Si -nuclear magnetic resonance SUPEKURU.

[Formula 16]



[0027]

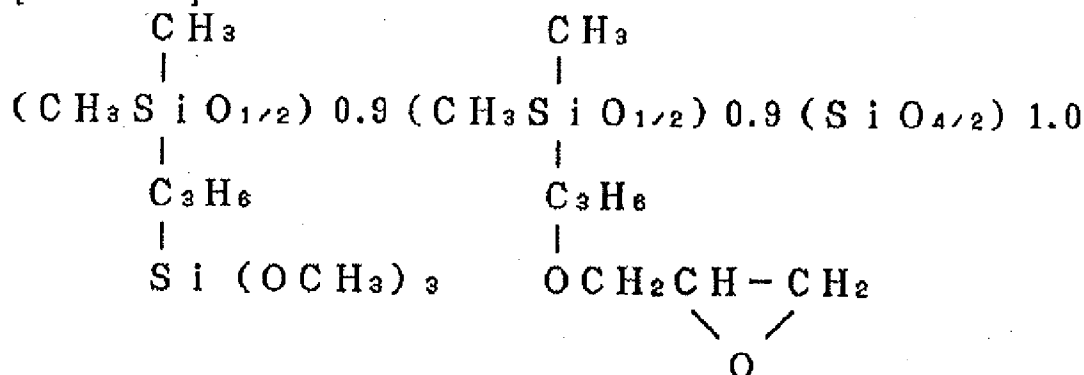
[Reference example 7] It is a formula to agitating equipment, a flowing-back condenser tube, and a 4 mouth flask with a thermometer. : [Formula 17]



Come out and the organopolysiloxane (0.92 % of the weight of content of 46 centipoise viscosity and a silicon atom absorbed water matter atom) expressed in toluene, Allyl trimethoxysilane of allyl glycidyl ether and the mol (to the number of mols of the silicon atom absorbed water matter atom in organopolysiloxane) The number of mols of the allyl group in allyl glycidyl ether and allyl trimethoxysilane is an excessive amount. It was made to react under existence of a chloroplatinic acid catalyst, and organopolysiloxane of yellow transparency was prepared. The viscosity of this organopolysiloxane is 200 centipoises. It was checked that this organopolysiloxane is organopolysiloxane expressed with a lower type by ^1H -nuclear-magnetic-resonance-spectrum, ^{13}C -nuclear-magnetic-resonance-spectrum, and

²⁹Si-nuclear magnetic resonance SUPEKURU.

[Formula 18]



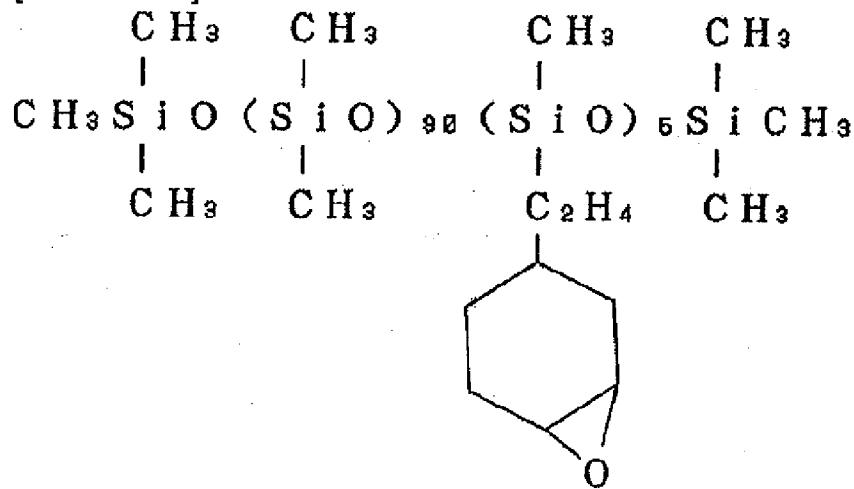
[0028]

[Work example 1] Phenol novolak resin (80 ** of softening temperature, hydroxyl equivalent 100) 35 weight section, Organopolysiloxane 3 weight section prepared by the reference example 1, fused-quartz powder 65 weight section, hexamethylenetetramine 4 weight section, and carnauba wax 1 weight section were kneaded with a 90 ** heating roller, and the hardenability resin composition was prepared. Subsequently, this was ground and transfer moulding was carried out for 3 minutes under the conditions of 175 **, and 70 kg / cm². Then, postcure of the hardening resin was carried out over 2 hours at 150 **. The various characteristics of the obtained hardening resin were shown in Table 1.

[0029]

[Comparative example 1] In Example 1, the hardenability resin composition was prepared like Example 1 except having used the organopolysiloxane expressed with a lower type instead of the organopolysiloxane prepared by the reference example 1. This was stiffened like Example 1. The various characteristics of the obtained hardening resin were shown in Table 1.

[Formula 19]



[0030]

[Comparative example 2] In Example 1, the hardenability resin composition was prepared like Example 1 except not using the organopolysiloxane prepared by the reference example 1. This was stiffened like Example 1. The various characteristics of the obtained hardening resin were shown in Table 1.

[0031]

[Table 1]

測 定 項 目	本 発 明	比 較 例	
	実施例 1	比較例 1	比較例 2
スパイラルフロー (インチ)	3 6	3 5	3 1
成形収縮率 (%)	0. 2 2	0. 2 0	0. 3 1
熱膨張率 ($\times 10^{-5}/^{\circ}\text{C}$)	1. 1 0	1. 0 5	1. 5 0
ガラス転移点 ($^{\circ}\text{C}$)	1 5 5	1 3 5	1 5 6
曲げ弾性率 (kg/mm^2)	1 2 1 0	1 2 5 0	1 3 5 0
吸水率 (%)	0. 4 2	0. 5 5	0. 5 1

[0032]

[Work example 2] $\text{CH}_3\text{SiO}_{3/40}$ mol of 2 unit %, and $\text{C}_6\text{H}_5(\text{CH}_3)\text{SiO}_{2/10}$ mol of 2 unit %, It consists of $\text{C}_6\text{H}_5\text{SiO}_{3/40}$ mol of 2 unit %, and $(\text{C}_6\text{H}_5)_2\text{SiO}_{2/10}$ mol of 2 unit %, Methylphenyl polysiloxane resin 13 weight section and cresol novolak epoxy resin (80 ** of softening temperatures) which contain the hydroxyl group linking directly to Si 5% of the weight The amount part of organopolysiloxane duplexs prepared by the reference example 2 to the silicone epoxy resin which consists of weight per epoxy equivalent 220 13 weight section, Fused-quartz powder 74 weight section, aluminum acetylacetonate 0.90 weight section, and carnauba wax 1 weight section were kneaded with a 90 ** heating roller, and the hardenability resin composition was obtained. Subsequently, this was ground and transfermold shaping was carried out for 2 minutes under the conditions of 175 **, and $70 \text{ kg} / \text{cm}^2$. Then, postcure of this hardening resin was carried out over 12 hours at 180 **. The various characteristics of the obtained hardening resin were shown in Table 2.

[0033]

[Work example 3] In Example 2, the hardenability resin composition was prepared like Example

2 except having used the organopolysiloxane prepared by the reference example 3 instead of the organopolysiloxane prepared by the reference example 2. This was stiffened like Example 2. The various characteristics of the obtained hardening resin were shown in Table 2.

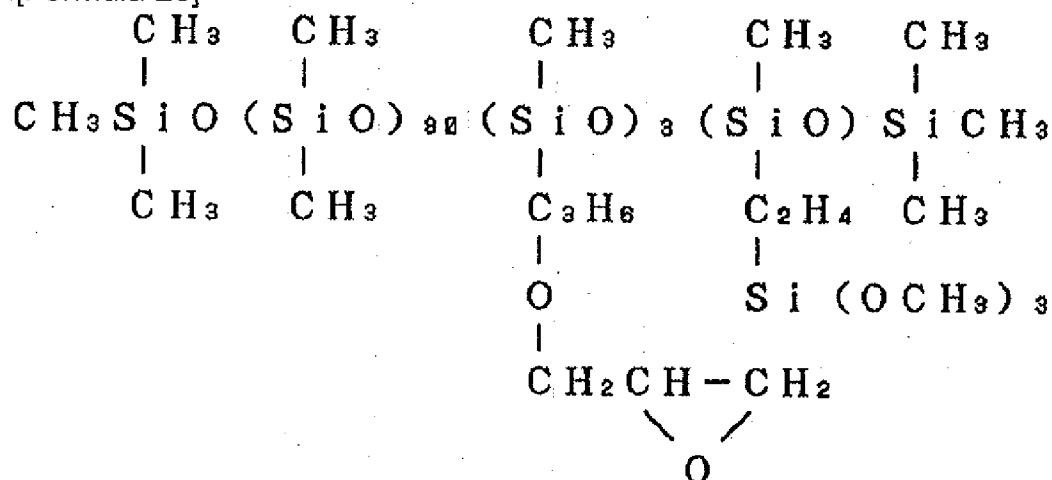
[0034]

[Work example 4] In Example 2, the hardenability resin composition was prepared like Example 2 except having used the organopolysiloxane prepared by the reference example 4 instead of the organopolysiloxane prepared by the reference example 2. This was stiffened like Example 2. The various characteristics of the obtained hardening resin were shown in Table 2.

[0035]

[Comparative example 3] In Example 2, the hardenability resin composition was prepared like Example 2 except having used the organopolysiloxane expressed with a lower type instead of the organopolysiloxane prepared by the reference example 2. This was stiffened like Example 2. The various characteristics of the obtained hardening resin were shown in Table 2.

[Formula 20]



[0036]

[Comparative example 4] In Example 2, the hardenability resin composition was prepared like Example 2 except not adding the organopolysiloxane prepared by the reference example 2. This was stiffened like Example 2. The various characteristics of the obtained hardening resin were shown in Table 2.

[0037]

[Table 2]

測定項目	本 発 明			比 較 例	
	実施例 2	実施例 3	実施例 4	比較例 3	比較例 4
スパイラルフロー(%)	46	43	41	40	35
成形収縮率 (%)	0.34	0.36	0.32	0.37	0.43
熱膨張率 ($\times 10^{-5}/^{\circ}\text{C}$)	2.32	2.25	2.20	2.90	2.75
ガラス転移点 ($^{\circ}\text{C}$)	175	173	176	151	170
曲げ弾性率 (kg/mm^2)	1100	1120	1140	1250	1520
吸水率 (%)	0.43	0.41	0.40	0.56	0.47
バリ (mm)	4	5	3	15	4
接着性	○	○	○	△	×
耐熱衝撃性	○	○	○	△	×
半田耐熱性	○	○	○	×	×

[0038]

[Work example 5]Thermosetting polyimide resin 35 weight section of a bismaleimide triazine type, Organopolysiloxane 4 weight section prepared by the reference example 5, fused-quartz powder 65 weight section, carnauba wax 1 weight section, and benzoic acid aluminum 0.32 weight section were kneaded with a 90 ** heating roller, and the hardenability resin composition was prepared. Subsequently, this was ground and transfermold shaping was carried out for 4 minutes under the conditions of 220 **, and $70 \text{ kg} / \text{cm}^2$. Then, postcure of this

hardening resin was carried out over 3 hours at 230 **. The various characteristics of the obtained hardening resin were shown in Table 3.

[0039]

[Comparative example 5] In Example 5, the hardenability resin composition was prepared like Example 5 except not using the organopolysiloxane prepared by the reference example 5. This was stiffened like Example 5. The various characteristics of this hardening resin were shown in Table 3.

[0040]

[Table 3]

測 定 項 目	本 発 明	比 較 例
	実施例 5	比較例 5
スパイラルフロー (インチ)	5 2	4 4
曲げ弾性率 (kg/mm ²)	9 8 0	1 2 2 0
接着性	○	×

[0041]

[Work example 6]Orthocresol-novolak-epoxy-resin (80 ** of softening temperature, weight per epoxy equivalent 220) 75 weight section, Fused silica 260 weight section, KARUNA bow wax 1 weight section, phenol novolak resin 35 weight section, triphenylphosphine 0.6 weight section, and organopolysiloxane 5 weight section prepared by the reference example 6 were kneaded with a 90 ** heating roller, and the hardenability resin composition was prepared.

Subsequently, this was ground and transfermold shaping was carried out for 3 minutes under the conditions of 150 **, and 70 kg / cm². Then, postcure of this hardening resin was carried out over 4 hours at 180 **. The various characteristics of the obtained hardening resin were shown in Table 4.

[0042]

[Work example 7]In Example 6, the hardenability resin composition was prepared like Example 6 except having used the organopolysiloxane prepared by the reference example 7 instead of the organopolysiloxane prepared by the reference example 6. This was stiffened like Example 6. The various characteristics of the obtained hardening resin were shown in Table 4.

[0043]

[Comparative example 6] In Example 6, the hardenability resin composition was prepared like Example 6 except not using the organopolysiloxane prepared by the reference example 6. This

was stiffened like Example 6. The various characteristics of the obtained hardening resin were shown in Table 4.

[0044]

[Table 4]

測定項目	本 発 明		比較例
	実施例 6	実施例 7	比較例 6
スパイラルフロー (インチ)	7 3	7 0	5 6
熱膨張率 ($\times 10^{-5}/^{\circ}\text{C}$)	2. 7 0	2. 6 5	2. 6 7
ガラス転移点 ($^{\circ}\text{C}$)	1 3 8	1 4 0	1 3 5
曲げ弾性率 (kg/mm^2)	1 1 5 0	1 2 1 0	1 4 2 0
吸水率 (%)	0. 4 9	0. 5 2	0. 5 3
接着性	○	△～○	×
耐熱衝撃性	○	○	×
半田耐熱性	○	○	×

[0045]

[Effect of the Invention] Since the hardenability resin composition of this invention has blended the organopolysiloxane which has an epoxy group joint organic group as a (B) ingredient, it has before hardening the feature that excel in mobility and the hardening resin produced by hardening this is excellent in flexibility and an adhesive property.

[Translation done.]

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TECHNICAL FIELD

[Industrial Application] This invention is excellent in mobility before hardening in detail about a hardenability resin composition, and after hardening is related with the hardenability resin composition used as hardening resin excellent in flexibility and an adhesive property.

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PRIOR ART

[Description of the Prior Art] Since the hardening resin produced by hardening this is excellent in mechanical properties, such as electrical properties, such as dielectric characteristics, volume resistivity, and electrical breakdown strength, or flexural strength, compressive strength, and impact strength, the hardenability resin composition is used for the electrical and electric equipment and sealing resin for electronic devices, adhesives, resin for FRP, etc. [0003] Generally, a hardenability resin composition has the upright hardening resin produced by hardening this, and Since [lacking in flexibility and] the rate of cure shrinkage at the time of hardening is large, If this is used as the electrical and electric equipment and sealing resin for electronic devices, in order to add big stress to the electrical and electric equipment and an electronic device, There was a problem of producing a crack in hardening resin itself, or destroying the electrical and electric equipment and an electronic device, and producing a crevice between the electrical and electric equipment and an electronic device, and hardening resin since the adhesive property over the electrical and electric equipment and electronic device of hardening resin is low. The coefficient of thermal expansion of the hardening resin produced by hardening this a hardenability resin composition to the coefficient of thermal expansion of the electrical and electric equipment and an electronic device Since it is large, If the resin seal electrical and electric equipment and an electronic device receive a thermo cycle, in order to add big stress to the electrical and electric equipment and an electronic device, There was a problem of producing a crack in hardening resin itself, or destroying the electrical and electric equipment and an electronic device, and producing a crevice between the electrical and electric equipment and an electronic device, and hardening resin since the adhesive property over the electrical and electric equipment and electronic device of hardening resin is low. When water invaded into the crevice between the crack or hardening resin produced by the cure shrinkage of a hardenability resin composition, or thermal expansion contraction, and the electrical and electric equipment and an electronic device, there was a

problem of promoting degradation of the resin seal electrical and electric equipment and an electronic device.

[0004] Thus, in the hardenability resin composition, in order to low-stress-ize hardening resin, the flexibility of hardening resin is raised, and the hardenability resin composition of the versatility which makes the adhesive property over the electrical and electric equipment and electronic device of hardening resin come to improve is proposed. For example, an epoxy resin, a methylphenyl polysiloxane, the organopolysiloxane that consists of bifunctional siloxane units (D unit) which have an epoxy group, and trifunctional siloxane units (T unit), The hardenability resin composition which consists of an inorganic filler and a catalyst for hardening (refer to JP,56-145942,A), . Have an epoxy resin, an epoxy group or an amino group and a hydroxyl group, or at least one basis that can be hydrolyzed, respectively. The organopolysiloxane which consists of monofunctional siloxane units (M unit) or trifunctional siloxane units (T unit), and bifunctional siloxane units (D unit), The hardenability resin composition which consists of a hardening agent and an inorganic filler (refer to JP,56-136816,A), An epoxy resin, phenol resin, the hardenability resin composition that becomes a chain side chain from the dimethylpolysiloxane which has an epoxy group and a polyoxyalkylene group (refer to JP,60-58425,A), And the hardenability resin composition (refer to JP,64-51467,A) which consists of hardening resin and silicone rubber spherical pulverized coal is proposed.

[0005] However, the hardenability resin composition proposed by JP,56-145942,A and JP,56-136816,A, Since the organopolysiloxane which makes bifunctional siloxane units (D unit) indispensable is used, There was a fault that the glass transition point (T_g) of the obtained hardening resin fell, and it was remarkable especially when using Loki Sun for the organopolysiloxane which consists of $2\text{SiO}_{2/a}2$ unit (D unit) which has an epoxy group (CH_3). The hardenability resin composition proposed by JP,60-58425,A, In order to use the dimethylpolysiloxane which has an epoxy group and a polyoxyalkylene group for a chain side chain, there was a fault that the obtained hardening resin had large absorptivity and it was not suitable as the electrical and electric equipment and sealing resin for electronic devices. The hardenability resin composition proposed by JP,64-51467,A, Although the flexibility of the hardening resin produced by hardening this was excellent, since the mobility of a hardenability resin composition was bad, there was a fault that the workability which carries out the resin seal of the electrical and electric equipment and the electronic device was bad, and this hardening resin had a still worse adhesive property over the electrical and electric equipment and an electronic device.

[0006]

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EFFECT OF THE INVENTION

[Effect of the Invention] Since the hardenability resin composition of this invention has blended the organopolysiloxane which has an epoxy group joint organic group as a (B) ingredient, it has before hardening the feature that excel in mobility and the hardening resin produced by hardening this is excellent in flexibility and an adhesive property.

[Translation done.]

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TECHNICAL PROBLEM

[Problem(s) to be Solved by the Invention] This invention persons reached this invention, as a result of trying hard wholeheartedly, in order to solve the above-mentioned problem.

[0007] That is, the purpose of this invention is excellent in mobility before hardening, and there is after hardening in providing the hardenability resin composition used as hardening resin excellent in flexibility and an adhesive property.

[Translation done.]

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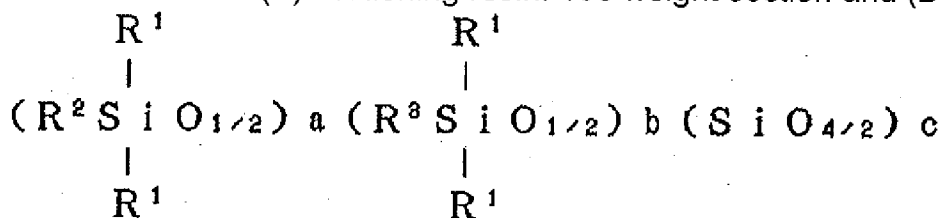
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OPERATION

[The means for solving a problem, and its operation]

This invention is (A) hardening resin. 100 weight section and (B) general formula :[Formula 2]



{R¹ is a monovalent hydrocarbon group except an alkenyl group among a formula, and R² is the monovalent hydrocarbon group or hydrogen atom except an alkenyl group, R³ is an epoxy group content organic group or an alkoxy silyl alkyl group, and a is 0 or a positive number, b is a positive number, c is a positive number, a/c is the number of 0-4, and b/c is the number of 0.05-4, and (a+b)/c is the number of 0.2-4. It is related with the hardenability resin composition which consists of 0.1 to organopolysiloxane 500 weight section which is expressed with}, and which has an epoxy group content organic group.

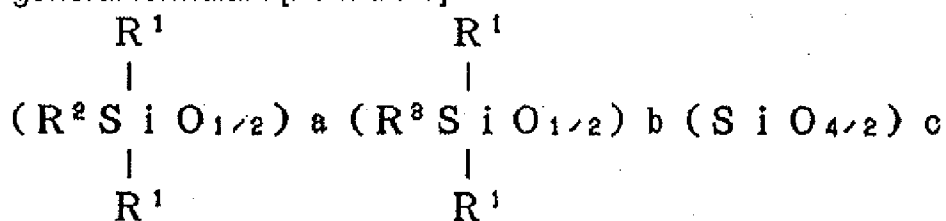
[0009] Hereafter, the hardenability resin composition of this invention is explained in detail.

[0010] (A) An ingredient is hardening resin which is a substrate of this invention. (A) As an ingredient, publicly known hardening resin can be used conventionally. (A) As an ingredient specifically, Phenol resin, formaldehyde resins, xylene resin, xylene-formaldehyde resin, ketone formaldehyde resins, furan resin, urea resin, imide resin, melamine resin, an alkyd resin, unsaturated polyester resin, aniline resin, sulfone amide resin, Silicone resin, epoxy resins, and these copolymer resin are illustrated, and it can be used combining one sort of the hardening resin of the above-mentioned illustration, or two sorts or more. It is preferred that it is hardening resin chosen from the group which consists of an epoxy resin, phenol resin, imide resin, and silicone resin especially as a (A) ingredient. The hardening resin etc. in which the

curing means hardens especially the (A) ingredient with the hardening resin hardened with high energy beams which it is not limited but are hardened with heat, such as hardening resin, ultraviolet rays, or radiation, and humidity are illustrated. At a room temperature, the (A) ingredient may be liquefied or may be hardening resin of which state of a solid state.

[0011](A) A hardening agent, a hardening accelerator, a bulking agent, a photosensitizer, higher-fatty-acid metal salt, an ester system wax, a plasticizer, etc. can be blended with an ingredient as ingredients other than the hardening resin of the above-mentioned illustration. As a hardening agent, specifically Organic acid, such as carboxylic acid and sulfonic acid, and the anhydride of those, an organic hydroxy compound, a silanol group, The amino compound of the organic silicon compound and the first class which have bases, such as an alkoxy group or a halogeno group, or the second class can be illustrated, and it can be used combining these kinds or two sorts or more. Specifically as a hardening accelerator, organophosphorus compounds, such as organic metallic compounds, such as the third class amine compound, aluminum, and a zirconium, and phosphine, a different ring type amine compound, a boron complex compound, organic ammonium salt, organic sulfonium salt, organic peroxide, etc. are illustrated. As a bulking agent, specifically, Glass fiber, asbestos, an alumina fiber, alumina, and silica. Fibrous fillers, such as the ceramic fiber used as an ingredient, a boron fiber, a zirconia fiber, silicon carbide fiber, a metal fiber, polyester fiber, an aramid fiber, a nylon fiber, a phenol fiber, and natural animals-and-plants textiles, fused silica, precipitation silica, fumed silica, pyrogenic silica, a zinc oxide, Calcination clay, carbon black, a glass bead, alumina, talc, calcium carbonate, clay, aluminium hydroxide, barium sulfate, a titanium dioxide, aluminium nitride, silicon carbide, magnesium oxide, beryllium oxide, kaolin, mica, Particulate matter-like bulking agents and these mixtures, such as zirconia, are illustrated.

[0012](B) An ingredient is an ingredient for raising the flexibility of the hardening resin produced by hardening the hardenability resin composition of this invention, and raising the adhesive property over the electrical and electric equipment and an electronic device, and is a general formula. : [Formula 3]



It is organopolysiloxane come out of and expressed. R^1 is a monovalent hydrocarbon group except an alkenyl group among an upper type, and specifically, A chloromethyl group, 3, 3, Aralkyl groups, such as aryl group; benzyls, such as alkyl group; phenyl groups, such as a methyl group, an ethyl group, a propyl group, a butyl group, a pentyl group, a hexyl group, and an octyl group, a tolyl group, and a xylyl group, and a phenethyl group; Substituted alkyl

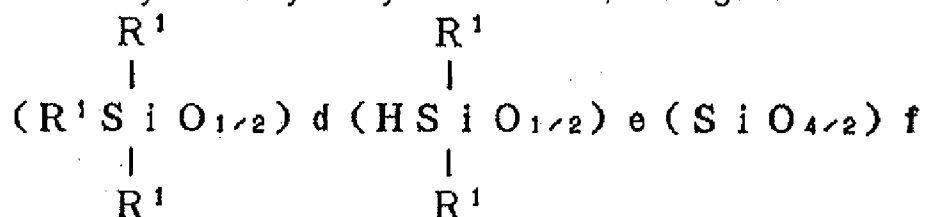
groups, such as 3-trifluoropropyl group, etc. are illustrated. R^2 is the monovalent hydrocarbon group or hydrogen atom except an alkenyl group, and as a monovalent hydrocarbon group except an alkenyl group specifically, A chloromethyl group, 3, 3, Aralkyl groups, such as aryl group; benzyls, such as alkyl group; phenyl groups, such as a methyl group, an ethyl group, a propyl group, a butyl group, a pentyl group, a hexyl group, and an octyl group, a tolyl group, and a xylyl group, and a phenethyl group; Substituted alkyl groups, such as 3-trifluoropropyl group, etc. are illustrated. R^3 is an epoxy group content organic group or an alkoxy silyl alkyl group, and as an epoxy group content organic group specifically, A glycidoxymethyl group, a glycidoxy propyl group, A glycidoxybutyl group, a 3,4-epoxycyclohexyl ethyl group, a 3,4-epoxycyclohexyl propyl group, a 3,4-epoxy NORUBORONE methyl ethyl group, a 2-(3,4-epoxy-3-methylcyclohexyl)-2-methylethyl group, etc. are illustrated, As an alkoxy silyl alkyl group, specifically, A trimethoxysilyl ethyl group, a triethoxy silyl ethyl group, a methyl dimethoxy silyl ethyl group, a trimethoxysilylpropyl group, a triethoxy silyl propyl group, a trimethoxysilyl butyl group, a methyl dimethoxy silyl propyl group, etc. are illustrated.

[0013]a is 0 or the positive number which shows the number of the monofunctional siloxane units (M unit) which do not have an epoxy group content organic group or an alkoxy silyl alkyl group among an upper type, b is a positive number which shows the number of the monofunctional siloxane units (M unit) which have an epoxy group content organic group or an alkoxy silyl alkyl group, c is a positive number which shows the number of tetrafunctional siloxane units (Q unit), each ratio and a/c are the number of the ranges of 0-4, and b/c is the number of the ranges of 0.05-4, and (a+b)/c is the number of the ranges of 0.2-4. Since monofunctional siloxane units (M unit) cannot have this [good] for four pieces to one tetrafunctional siloxane units (Q unit) and compatibility is excellent in the (B) ingredient to the (A) ingredient in this invention, It is because the monofunctional siloxane units (M unit) which have an epoxy group content organic group or an alkoxy silyl alkyl group need to be at least 0.05 piece to one tetrafunctional siloxane units (Q unit). (B) Although organopolysiloxane of an ingredient is liquefied at a room temperature, or it is a solid state and the molecular weight in particular is not limited, since compatibility with the (A) ingredient is good, it is preferred that it is the range of 500-500,000.

[0014]In this invention, organopolysiloxane of the (B) ingredient, Since it has an epoxy group content organic group or an alkoxy silyl alkyl group, (A) Since the flexibility of the hardening resin obtained by being incorporated into the resin matrix of an ingredient improves and the (B) ingredient consists of a SiO_2 unit (Q unit), the glass transition point (T_g) of the obtained hardening resin does not get worse, but has the advantage of excelling in heat resistance and fire retardancy.

[0015]Organopolysiloxane of the above-mentioned (B) ingredient is the bottom of existence of

the catalyst for a hydrosilylation reaction, and a general formula, for example. : [Formula 4]



{R¹ is a monovalent hydrocarbon group except an alkenyl group among a formula, d is 0 or a positive number, e is a positive number, f is a positive number, d/f is the number of 0-4, and e/f is the number of 0.05-4, and (d+e)/f is the number of 0.2-4. It can obtain by carrying out the addition reaction of the alkoxy silylalkene of the organic compound which has organopolysiloxane and the epoxy group which have a silicon atom absorbed water matter atom, and aliphatic unsaturated bonds which are expressed with}, and an arbitrary dose.

[0016](B) The loadings of an ingredient are 0.1 - 500 weight section to (A) ingredient 100 weight section.

It is 0.5 to 100 weight section preferably.

This is because there is no effect that the loadings of the (B) ingredient are less than 0.1 weight sections to (A) ingredient 100 weight section in low stress-ization of hardening resin.

It is for the mechanical strength of hardening resin to fall remarkably, if 500 weight sections are exceeded.

[0017]The hardenability resin composition of this invention is obtained by mixing the (A) ingredient and the (B) ingredient uniformly. (A) The method in particular of mixing an ingredient and the (B) ingredient is not limited, but specifically, (A) After blending and carrying out the premix of the (B) ingredient at the time of the method of blending the (B) ingredient with an ingredient directly, and (A) ingredient preparation, the method of blending additive agents, such as a bulking agent, with the (A) ingredient and the method of blending with the (B) ingredient various kinds of additive agents blended with the (A) ingredient one by one are illustrated. (A) Although there is no limitation and a mixed device is chosen by the gestalt of versatility [ingredient / the (A) ingredient or / (B)], such as a liquid, a solid state, and powder, especially as a mixed device which mixes an ingredient and the (B) ingredient, Specifically, the continuous mixer of one axis or two axes, 2 rolls, a loss mixer, and a kneader mixer are illustrated.

[0018]Since the hardenability resin composition of this invention is excellent in mobility before hardening, it can be used by methods, such as a transfer mold, injection molding, potting, casting, powder coating, dip coating, and dropping. Since after hardening becomes hardening resin excellent in flexibility and an adhesive property, the hardenability resin composition of this invention can be used for the hardenability resin composition for closing the electrical and

electric equipment and an electronic device, adhesives, etc.

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EXAMPLE

[Example] Hereafter, an example explains this invention. The value of viscosity is a value measured in 25 ** among an example, and the various characteristics of a hardenability resin composition and hardening resin were measured by the method shown below.

[0020] O Spiral flow : it measured by the method according to an EMMI standard.

O Molding shrinkage : It measured by the method according to JIS-K-6911.

O Coefficient of thermal expansion : The hardening resin fabricated to 5mmx5mmx16mm was measured using the thermal expansion meter (DL-7000 by a vacuum science-and-engineering company). The value of a coefficient of thermal expansion is a value from a room temperature to a glass transition point.

O Glass transition point (Tg) : it measured by coefficient-of-thermal-expansion measurement.

O Rate of bending flexibility : It measured by the method according to JIS-K-6911.

O Water absorption : After making into the temperature of 121 **, and 100% of humidity the hardening resin fabricated to 2 inch x 0.5 inch x 0.25 inch and humidifying it for 20 hours, it asked by measuring the weight increment of the hardening resin.

O Barricade : The 20-micrometer-deep slot was used and barricade length was measured.

O Adhesive property : After inserting the hardenability resin composition into 42 alloy board and the copper plate and stiffening this, the adhesive property at the time of removing hardening resin was judged by O (good) and x (poor).

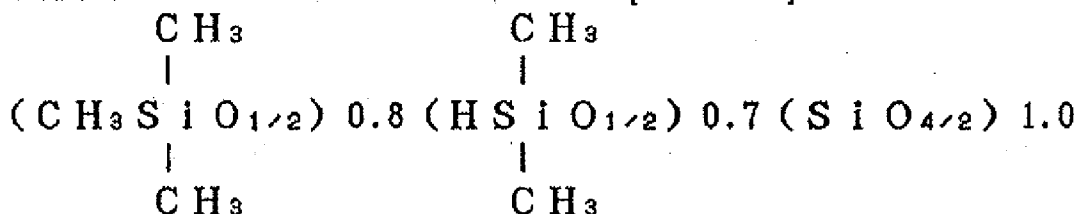
O Thermal shock resistance : 20 resin seal semiconductor devices (chip size 36 mm², and package 2.0 mm in thickness) are fabricated, This was performed at -196 **<-->+150 **, the heat cycle test was done in the cycle for 1 minute, the sealing resin surface was observed with the stereoscopic microscope after 150 cycles, and the case where they were ** and 11 pieces or more about the case where they are O and 6-10 pieces about the case where the number of the mold goods in which the crack went into the surface is five or less pieces was made into x.

O Solder heat resistance : 20 resin seal semiconductor devices (chip size 36 mm², and

package 2.0 mm in thickness) were fabricated, this was promptly immersed in a 240 ** solder bath after 72-hour neglect under 85%RH conditions at 85 ** for 1 hour, and the sealing resin surface was observed with the stereoscopic microscope. The case where they were ** and 11 pieces or more about the case where they are O and 6-10 pieces about the case where the number of the mold goods in which the crack went into the surface is five or less pieces was made into x.

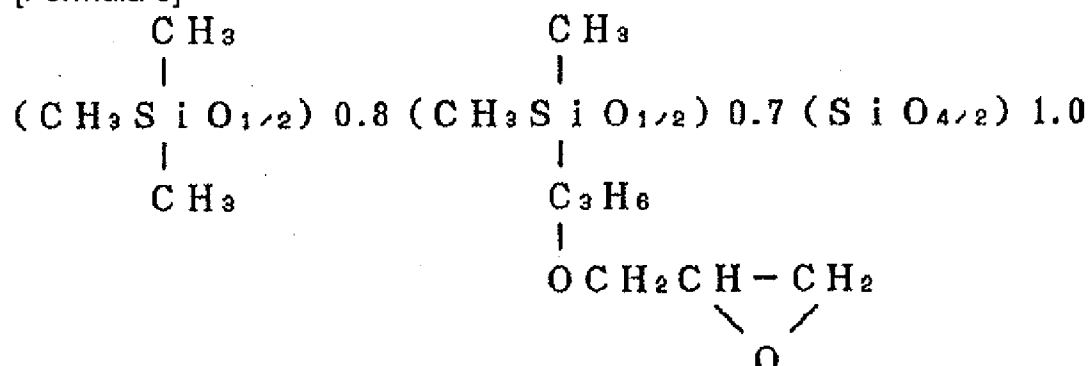
[0021]

[Reference example 1] It is a formula to agitating equipment, a flowing-back condenser tube, and a 4 mouth flask with a thermometer. : [Formula 5]



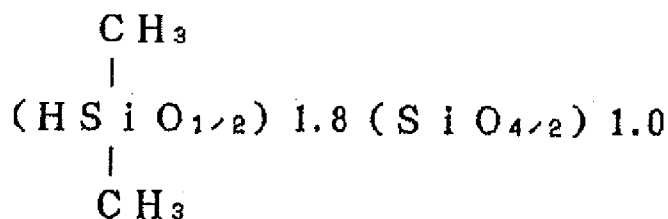
Come out and the organopolysiloxane (0.40 % of the weight of content of 105 centipoise viscosity and a silicon atom absorbed water matter atom) expressed in toluene, Allyl glycidyl ether (the number of mols of the allyl group in allyl glycidyl ether is an excessive amount to the number of mols of the silicon atom absorbed water matter atom in organopolysiloxane.) was made to react under existence of a chloroplatinic acid catalyst, and transparent brown organopolysiloxane was prepared. The viscosity of this organopolysiloxane was 520 centipoises and the weight per epoxy equivalent was 420. It was checked that this organopolysiloxane is organopolysiloxane expressed with a lower type by ^1H -nuclear-magnetic-resonance-spectrum, ^{13}C -nuclear-magnetic-resonance-spectrum, and ^{29}Si -nuclear magnetic resonance SUPEKURU.

[Formula 6]



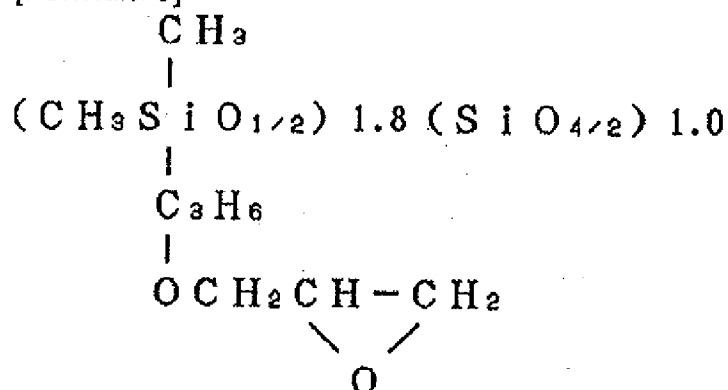
[0022]

[Reference example 2] It is a formula to agitating equipment, a flowing-back condenser tube, and a 4 mouth flask with a thermometer. : [Formula 7]



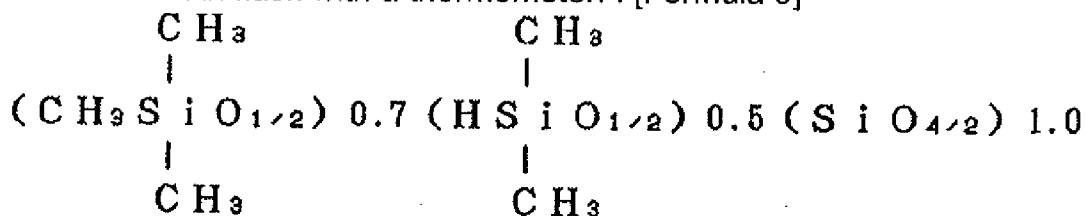
Come out and the organopolysiloxane (0.92 % of the weight of content of 46 centipoise viscosity and a silicon atom absorbed water matter atom) expressed in toluene, Allyl glycidyl ether (the number of mols of the allyl group in allyl glycidyl ether is an excessive amount to the number of mols of the silicon atom absorbed water matter atom in organopolysiloxane.) was made to react under existence of a chloroplatinic acid catalyst, and transparent brown organopolysiloxane was prepared. The viscosity of this organopolysiloxane was 610 centipoises and the weight per epoxy equivalent was 370. It was checked that this organopolysiloxane is organopolysiloxane expressed with a lower type by ^1H -nuclear-magnetic-resonance-spectrum, ^{13}C -nuclear-magnetic-resonance-spectrum, and ^{29}Si -nuclear magnetic resonance SUPEKURU.

[Formula 8]



[0023]

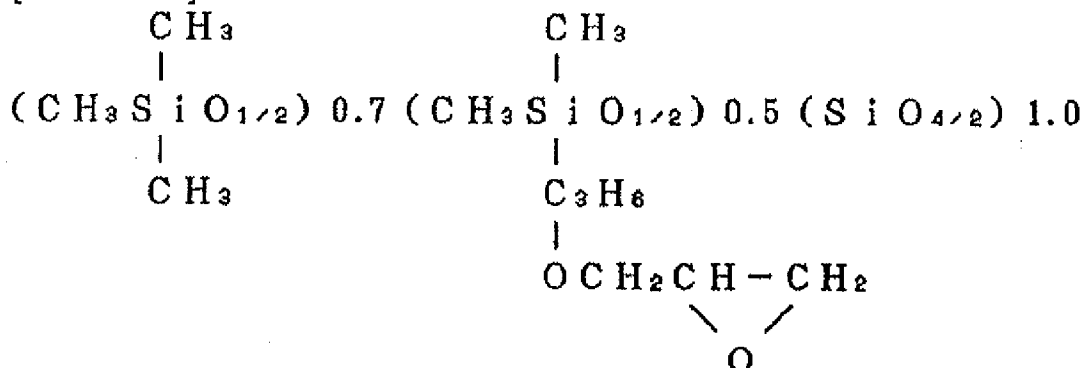
[Reference example 3] It is a formula to agitating equipment, a flowing-back condenser tube, and a 4 mouth flask with a thermometer. : [Formula 9]



Come out and the consistency organopolysiloxane (0.33 % of the weight of content of a silicon atom absorbed water matter atom) expressed in toluene, Allyl glycidyl ether (the number of mols of the allyl group in allyl glycidyl ether is an excessive amount to the number of mols of the silicon atom absorbed water matter atom in organopolysiloxane.) was made to react under

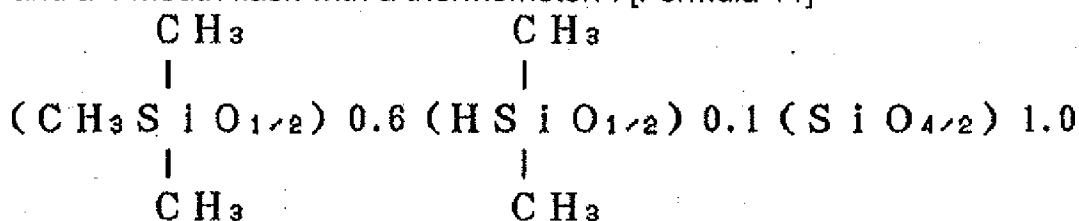
existence of a chloroplatinic acid catalyst, and organopolysiloxane of the brown shape of a translucent starch syrup was prepared. The weight per epoxy equivalent of this organopolysiloxane was 1100. It was checked that this organopolysiloxane is organopolysiloxane expressed with a lower type by ^1H -nuclear-magnetic-resonance-spectrum, ^{13}C -nuclear-magnetic-resonance-spectrum, and ^{29}Si -nuclear magnetic resonance SUPEKURU.

[Formula 10]



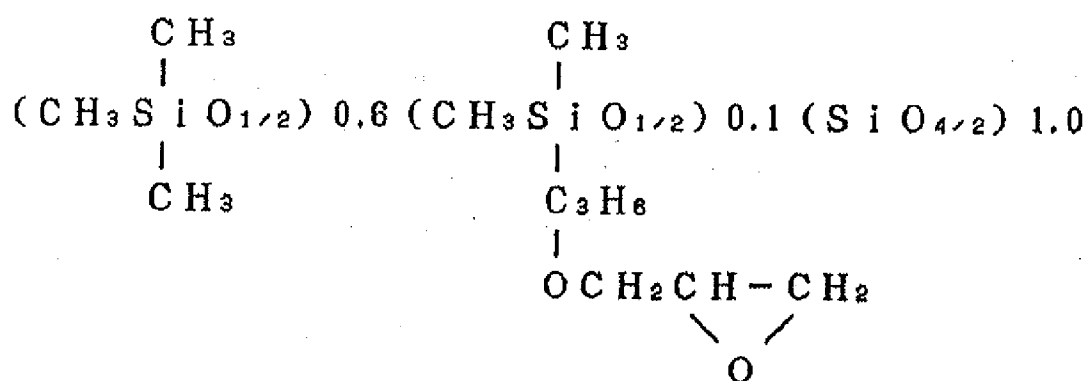
[0024]

[Reference example 4] It is a formula to agitating equipment, a flowing-back condenser tube, and a 4 mouth flask with a thermometer. : [Formula 11]



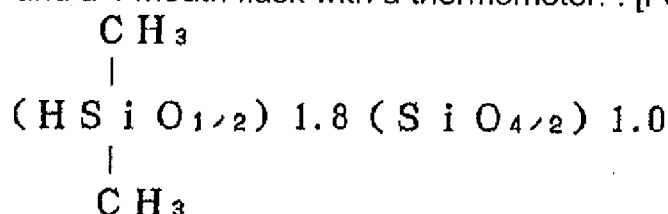
Come out and to 68% of the weight of the toluene solution of the organopolysiloxane (0.09 % of the weight of content of a silicon atom absorbed water matter atom) expressed. Allyl glycidyl ether (the number of mols of the allyl group in allyl glycidyl ether is an excessive amount to the number of mols of the silicon atom absorbed water matter atom in organopolysiloxane.) was made to react under existence of a chloroplatinic acid catalyst, and organopolysiloxane of the brown shape of a translucent starch syrup was prepared. The weight per epoxy equivalent of this organopolysiloxane was 1290. It was checked that this organopolysiloxane is organopolysiloxane expressed with a lower type by ^1H -nuclear-magnetic-resonance-spectrum, ^{13}C -nuclear-magnetic-resonance-spectrum, and ^{29}Si -nuclear magnetic resonance SUPEKURU.

[Formula 12]



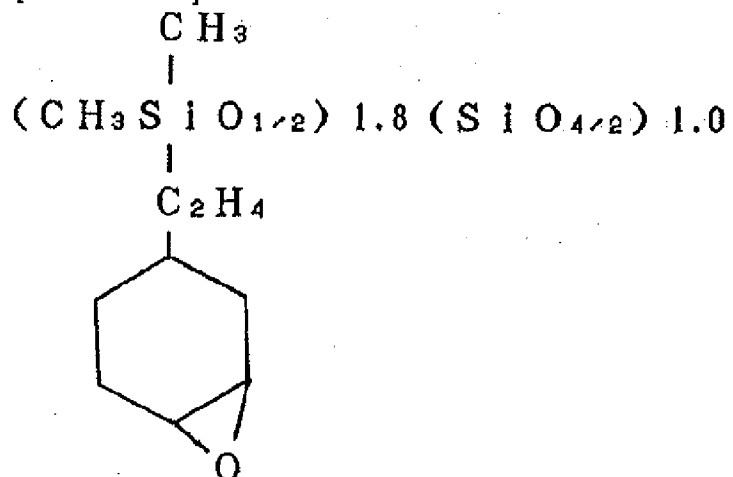
[0025]

[Reference example 5] It is a formula to agitating equipment, a flowing-back condenser tube, and a 4 mouth flask with a thermometer. : [Formula 13]



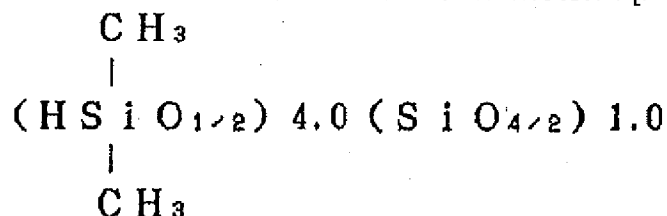
Come out and the organopolysiloxane (0.92 % of the weight of content of 46 centipoise viscosity and a silicon atom absorbed water matter atom) expressed in toluene, 1,2-epoxy-4-vinylcyclohexane (to the number of mols of the silicon atom absorbed water matter atom in organopolysiloxane) The number of mols of the vinyl group in 1,2-epoxy-4-vinylcyclohexane is an excessive amount. It was made to react under existence of a chloroplatinic acid catalyst, and transparent brown organopolysiloxane was prepared. The viscosity of this organopolysiloxane was 520 centipoises and the weight per epoxy equivalent was 230. It was checked that this organopolysiloxane is organopolysiloxane expressed with a lower type by ^1H -nuclear-magnetic-resonance-spectrum, ^{13}C -nuclear-magnetic-resonance-spectrum, and ^{29}Si -nuclear magnetic resonance SUPEKURU.

[Formula 14]



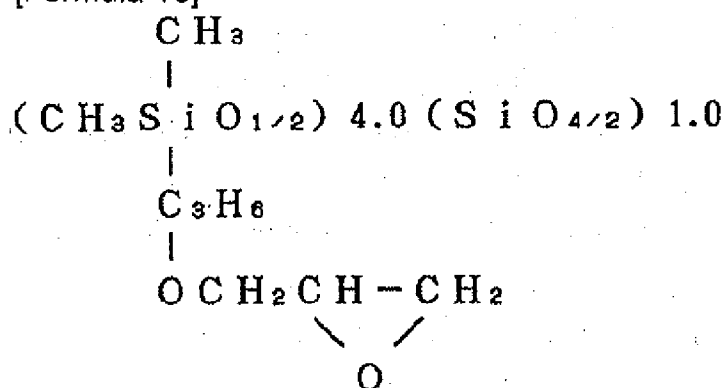
[0026]

[Reference example 6] It is a formula to agitating equipment, a flowing-back condenser tube, and a 4 mouth flask with a thermometer. : [Formula 15]



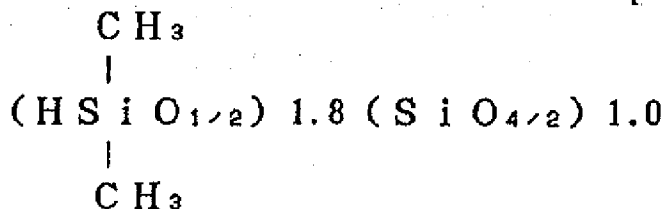
Come out and the organopolysiloxane (1.22 % of the weight of content of 190 ** of boiling points, and a silicon atom absorbed water matter atom) expressed in toluene, Allyl glycidyl ether (the number of mols of the allyl group in allyl glycidyl ether is an excessive amount to the number of mols of the silicon atom absorbed water matter atom in organopolysiloxane.) was made to react under existence of a chloroplatinic acid catalyst, and transparent brown organopolysiloxane was prepared. The viscosity of this organopolysiloxane was 54 centipoises and the weight per epoxy equivalent was 205. It was checked that this organopolysiloxane is organopolysiloxane expressed with a lower type by ^1H -nuclear-magnetic-resonance-spectrum, ^{13}C -nuclear-magnetic-resonance-spectrum, and ^{29}Si -nuclear magnetic resonance SUPEKURU.

[Formula 16]



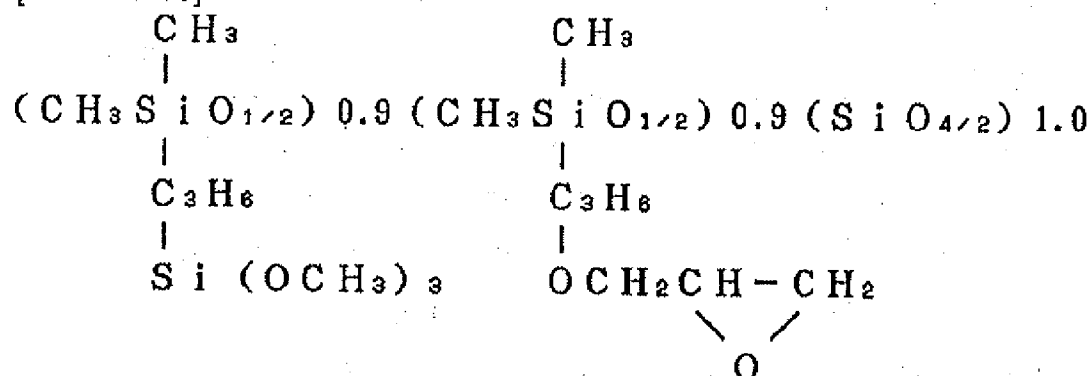
[0027]

[Reference example 7] It is a formula to agitating equipment, a flowing-back condenser tube, and a 4 mouth flask with a thermometer. : [Formula 17]



Come out and the organopolysiloxane (0.92 % of the weight of content of 46 centipoise viscosity and a silicon atom absorbed water matter atom) expressed in toluene, Allyl trimethoxysilane of allyl glycidyl ether and the mol (to the number of mols of the silicon atom absorbed water matter atom in organopolysiloxane) The number of mols of the allyl group in allyl glycidyl ether and allyl trimethoxysilane is an excessive amount. It was made to react under existence of a chloroplatinic acid catalyst, and organopolysiloxane of yellow transparence was prepared. The viscosity of this organopolysiloxane is 200 centipoises. It was checked that this organopolysiloxane is organopolysiloxane expressed with a lower type by ^1H -nuclear-magnetic-resonance-spectrum, ^{13}C -nuclear-magnetic-resonance-spectrum, and ^{29}Si -nuclear magnetic resonance SUPEKURU.

[Formula 18]



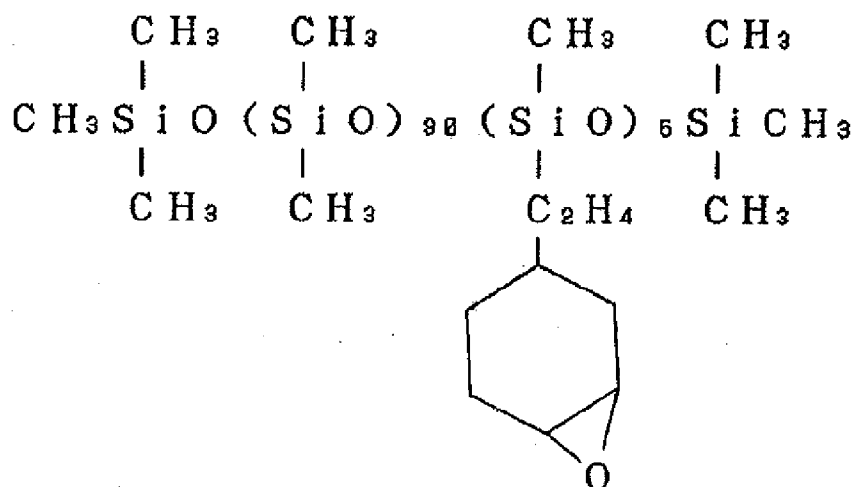
[0028]

[Work example 1] Phenol novolak resin (80 ** of softening temperature, hydroxyl equivalent 100) 35 weight section, Organopolysiloxane 3 weight section prepared by the reference example 1, fused-quartz powder 65 weight section, hexamethylenetetramine 4 weight section, and carnauba wax 1 weight section were kneaded with a 90 ** heating roller, and the hardenability resin composition was prepared. Subsequently, this was ground and transfer moulding was carried out for 3 minutes under the conditions of 175 **, and 70 kg / cm². Then, postcure of the hardening resin was carried out over 2 hours at 150 **. The various characteristics of the obtained hardening resin were shown in Table 1.

[0029]

[Comparative example 1] In Example 1, the hardenability resin composition was prepared like Example 1 except having used the organopolysiloxane expressed with a lower type instead of the organopolysiloxane prepared by the reference example 1. This was stiffened like Example 1. The various characteristics of the obtained hardening resin were shown in Table 1.

[Formula 19]



[0030]

[Comparative example 2] In Example 1, the hardenability resin composition was prepared like Example 1 except not using the organopolysiloxane prepared by the reference example 1. This was stiffened like Example 1. The various characteristics of the obtained hardening resin were shown in Table 1.

[0031]

[Table 1]

測定項目	本発明	比較例	
	実施例 1	比較例 1	比較例 2
スパイラルフロー (インチ)	3.6	3.5	3.1
成形収縮率 (%)	0.22	0.20	0.31
熱膨張率 ($\times 10^{-6}/^{\circ}\text{C}$)	1.10	1.05	1.50
ガラス転移点 ($^{\circ}\text{C}$)	155	135	156
曲げ弾性率 (kg/mm^2)	1210	1250	1350
吸水率 (%)	0.42	0.55	0.51

[0032]

[Work example 2] $\text{CH}_3\text{SiO}_{3/40}$ mol of 2 unit %, and $\text{C}_6\text{H}_5(\text{CH}_3)\text{SiO}_{2/10}$ mol of 2 unit %, It consists of $\text{C}_6\text{H}_5\text{SiO}_{3/40}$ mol of 2 unit %, and $(\text{C}_6\text{H}_5)_2\text{SiO}_{2/10}$ mol of 2 unit %, Methylphenyl

polysiloxane resin 13 weight section and cresol novolak epoxy resin (80 ** of softening temperatures) which contain the hydroxyl group linking directly to Si 5% of the weight The amount part of organopolysiloxane duplexs prepared by the reference example 2 to the silicone epoxy resin which consists of weight per epoxy equivalent 220 13 weight section, Fused-quartz powder 74 weight section, aluminum acetylacetonate 0.90 weight section, and carnauba wax 1 weight section were kneaded with a 90 ** heating roller, and the hardenability resin composition was obtained. Subsequently, this was ground and transfermold shaping was carried out for 2 minutes under the conditions of 175 **, and 70 kg / cm². Then, postcure of this hardening resin was carried out over 12 hours at 180 **. The various characteristics of the obtained hardening resin were shown in Table 2.

[0033]

[Work example 3] In Example 2, the hardenability resin composition was prepared like Example 2 except having used the organopolysiloxane prepared by the reference example 3 instead of the organopolysiloxane prepared by the reference example 2. This was stiffened like Example 2. The various characteristics of the obtained hardening resin were shown in Table 2.

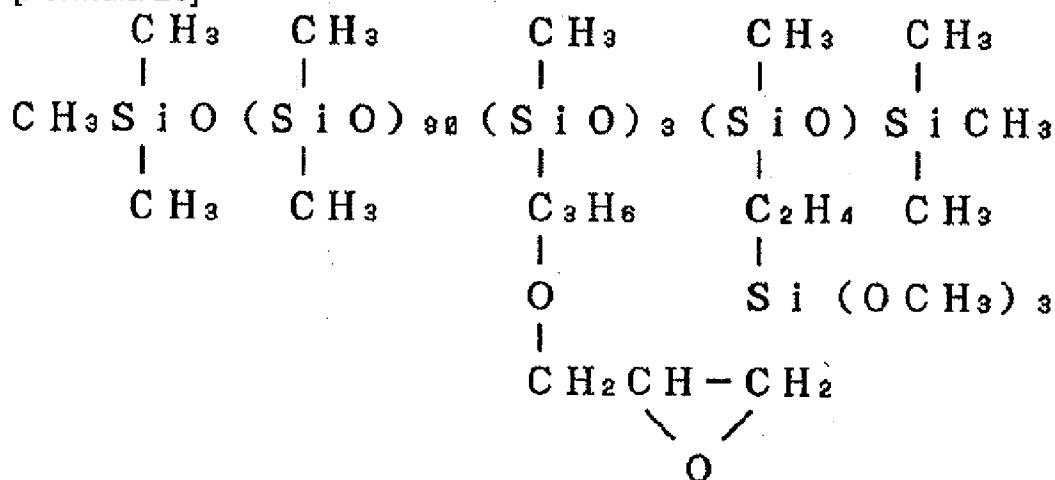
[0034]

[Work example 4] In Example 2, the hardenability resin composition was prepared like Example 2 except having used the organopolysiloxane prepared by the reference example 4 instead of the organopolysiloxane prepared by the reference example 2. This was stiffened like Example 2. The various characteristics of the obtained hardening resin were shown in Table 2.

[0035]

[Comparative example 3] In Example 2, the hardenability resin composition was prepared like Example 2 except having used the organopolysiloxane expressed with a lower type instead of the organopolysiloxane prepared by the reference example 2. This was stiffened like Example 2. The various characteristics of the obtained hardening resin were shown in Table 2.

[Formula 20]



[0036]

[Comparative example 4] In Example 2, the hardenability resin composition was prepared like Example 2 except not adding the organopolysiloxane prepared by the reference example 2. This was stiffened like Example 2. The various characteristics of the obtained hardening resin were shown in Table 2.

[0037]

[Table 2]

測定項目	本発明			比較例	
	実施例 2	実施例 3	実施例 4	比較例 3	比較例 4
スパイラルフロー(1/f)	46	43	41	40	35
成形収縮率 (%)	0.34	0.36	0.32	0.37	0.43
熱膨張率 ($\times 10^{-5}/^{\circ}\text{C}$)	2.32	2.25	2.20	2.90	2.75
ガラス転移点 ($^{\circ}\text{C}$)	175	173	176	151	170
曲げ弾性率 (kg/mm^2)	1100	1120	1140	1250	1520
吸水率 (%)	0.43	0.41	0.40	0.56	0.47
バリ (mm)	4	5	3	15	4
接着性	○	○	○	△	×
耐熱衝撃性	○	○	○	△	×
半田耐熱性	○	○	○	×	×

[0038]

[Work example 5]Thermosetting polyimide resin 35 weight section of a bismaleimide triazine type, Organopolysiloxane 4 weight section prepared by the reference example 5, fused-quartz powder 65 weight section, carnauba wax 1 weight section, and benzoic acid aluminum 0.32 weight section were kneaded with a 90 ° heating roller, and the hardenability resin composition was prepared. Subsequently, this was ground and transfermold shaping was carried out for 4 minutes under the conditions of 220 °, and 70 kg / cm². Then, postcure of this hardening resin was carried out over 3 hours at 230 °. The various characteristics of the obtained hardening resin were shown in Table 3.

[0039]

[Comparative example 5] In Example 5, the hardenability resin composition was prepared like Example 5 except not using the organopolysiloxane prepared by the reference example 5. This was stiffened like Example 5. The various characteristics of this hardening resin were shown in Table 3.

[0040]

[Table 3]

測 定 項 目	本 発 明	比 較 例
	実 施 例 5	比 較 例 5
スパイラルフロー (インチ)	5 2	4 4
曲げ弾性率 (kg/mm ²)	9 8 0	1 2 2 0
接着性	○	×

[0041]

[Work example 6]Orthocresol-novolak-epoxy-resin (80 ° of softening temperature, weight per epoxy equivalent 220) 75 weight section, Fused silica 260 weight section, KARUNA bow wax 1 weight section, phenol novolak resin 35 weight section, triphenylphosphine 0.6 weight section, and organopolysiloxane 5 weight section prepared by the reference example 6 were kneaded with a 90 ° heating roller, and the hardenability resin composition was prepared. Subsequently, this was ground and transfermold shaping was carried out for 3 minutes under the conditions of 150 °, and 70 kg / cm². Then, postcure of this hardening resin was carried out over 4 hours at 180 °. The various characteristics of the obtained hardening resin were shown in Table 4.

[0042]

[Work example 7] In Example 6, the hardenability resin composition was prepared like Example 6 except having used the organopolysiloxane prepared by the reference example 7 instead of the organopolysiloxane prepared by the reference example 6. This was stiffened like Example 6. The various characteristics of the obtained hardening resin were shown in Table 4.

[0043]

[Comparative example 6] In Example 6, the hardenability resin composition was prepared like Example 6 except not using the organopolysiloxane prepared by the reference example 6. This was stiffened like Example 6. The various characteristics of the obtained hardening resin were shown in Table 4.

[0044]

[Table 4]

測 定 項 目	本 発 明		比較例
	実施例 6	実施例 7	比較例 6
スパイラルフロー (インチ)	7 3	7 0	5 6
熱膨張率 ($\times 10^{-5}/^{\circ}\text{C}$)	2. 7 0	2. 6 5	2. 6 7
ガラス転移点 ($^{\circ}\text{C}$)	1 3 8	1 4 0	1 3 5
曲げ弾性率 (kg/mm^2)	1 1 5 0	1 2 1 0	1 4 2 0
吸水率 (%)	0. 4 9	0. 5 2	0. 5 3
接着性	○	△～○	×
耐熱衝撃性	○	○	×
半田耐熱性	○	○	×

[0045]

[Translation done.]

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C 0 8 L 63/00	N J S	8830-4 J		
	N J W	8830-4 J		
	N K A	8830-4 J		
	N K B	8830-4 J		

審査請求 未請求 請求項の数2(全13頁)

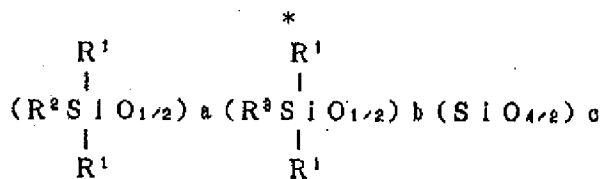
(21)出願番号	特願平4-128081	(71)出願人	000110077 東レ・ダウコーニング・シリコン株式会社 東京都中央区日本橋室町2丁目3番16号
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(54)【発明の名称】 硬化性樹脂組成物

(57)【要約】

【目的】 流動性に優れ、硬化して可撓性および接着性に優れた硬化樹脂となる硬化性樹脂組成物を提供することにある。

*【構成】 (A)硬化性樹脂100重量部および(B)一般式:
【化1】



{式中、R¹はアルケニル基を除く一価炭化水素基であり、R²はアルケニル基を除く一価炭化水素基または水素原子であり、R³はエポキシ基含有有機基またはアルコキシシリルアルキル基であり、またaは0または正数であり、bは正数であり、cは正数であり、a/cは0

～4の数であり、b/cは0.05～4の数であり、かつ(a+b)/cは0.2～4の数である。}で表される、エポキシ基含有有機基を有するオルガノポリシロキサン0.1～500重量部からなる硬化性樹脂組成物。

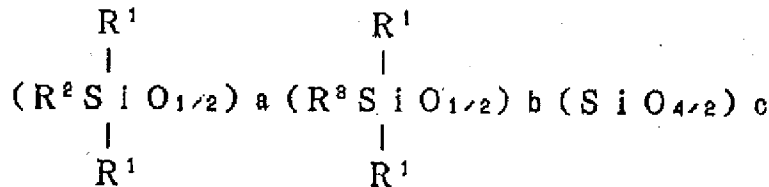
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【特許請求の範囲】

【請求項1】 (A)硬化性樹脂

100重量部



【式中、 R^1 はアルケニル基を除く一価炭化水素基であり、 R^2 はアルケニル基を除く一価炭化水素基または水素原子であり、 R^3 はエポキシ基含有有機基またはアルコキシシリルアルキル基であり、また a は0または正数であり、 b は正数であり、 c は正数であり、 a/c は0~4の数であり、 b/c は0.05~4の数であり、かつ $(a+b)/c$ は0.2~4の数である。】で表される、エポキシ基含有有機基を有するオルガノポリシロキサン

0.1~500重量部

からなる硬化性樹脂組成物。

【請求項2】 (A)成分の硬化性樹脂が、エポキシ樹脂、フェノール樹脂、イミド樹脂およびシリコン樹脂からなる群から選択される硬化性樹脂である請求項1記載の硬化性樹脂組成物。

【発明の詳細な説明】

【0001】

【産業上の利用分野】本発明は、硬化性樹脂組成物に関し、詳しくは、硬化前は流動性が優れ、硬化後は可撓性および接着性が優れた硬化樹脂となる、硬化性樹脂組成物に関する。

【0002】

【従来の技術】硬化性樹脂組成物は、これを硬化して得られた硬化樹脂が、誘電特性、体積抵抗率および絶縁破壊強度等の電気的特性あるいは曲げ強度、圧縮強度および衝撃強度等の機械的特性に優れているため、電気・電子素子用封止樹脂、接着剤、FRP用樹脂等を使用されている。

【0003】硬化性樹脂組成物は、これを硬化して得られた硬化樹脂が一般に剛直で可撓性に乏しく、また硬化時の硬化収縮率が大きいと、これを電気・電子素子用封止樹脂として使用すると、電気・電子素子に対して大きな応力が加わるため、硬化樹脂自体に亀裂を生じたり、あるいは電気・電子素子を破壊したり、また硬化樹脂の電気・電子素子に対する接着性が低いと、電気・電子素子と硬化樹脂との間に隙間を生じたりするという問題があった。また、硬化性樹脂組成物は、これを硬化して得られた硬化樹脂の熱膨張係数が電気・電子素子の熱膨張係数に対して大きいと、樹脂封止電気・電子素子がヒートサイクルを受けると、電気・電子素子に対して大きな応力が加わるため、硬化樹脂自体に亀裂を生じ

*および、(B)一般式：

【化1】

たり、あるいは電気・電子素子を破壊したり、また硬化樹脂の電気・電子素子に対する接着性が低いと、電気・電子素子と硬化樹脂との間に隙間を生じたりするという問題があった。さらに、硬化性樹脂組成物の硬化収縮あるいは熱膨張収縮によって生じた亀裂または硬化樹脂と電気・電子素子との隙間に水が侵入することにより、樹脂封止電気・電子素子の劣化を促進するという問題があった。

【0004】このように、硬化性樹脂組成物において、硬化樹脂を低応力化するために硬化樹脂の可撓性を向上させたり、また硬化樹脂の電気・電子素子に対する接着性を向上させてなる種々の硬化性樹脂組成物が提案されている。例えば、エポキシ樹脂、メチルフェニルポリシロキサン、エポキシ基を有する二官能性シロキサン単位(D単位)と三官能性シロキサン単位(T単位)からなるオルガノポリシロキサン、無機質充填剤および硬化用触媒からなる硬化性樹脂組成物(特開昭56-145942号公報参照)、エポキシ樹脂、エポキシ基またはアミノ基および水酸基または加水分解可能な基をそれぞれ少なくとも1個有する、一官能性シロキサン単位(M単位)または三官能性シロキサン単位(T単位)と二官能性シロキサン単位(D単位)からなるオルガノポリシロキサン、硬化剤および無機質充填剤からなる硬化性樹脂組成物(特開昭56-136816号公報参照)、エポキシ樹脂、フェノール樹脂、分子鎖側鎖にエポキシ基とポリオキシアルキレン基を有するジメチルポリシロキサンからなる硬化性樹脂組成物(特開昭60-58425号公報参照)、および硬化性樹脂とシリコンゴム球状微粉体からなる硬化性樹脂組成物(特開昭64-51467号公報参照)が提案されている。

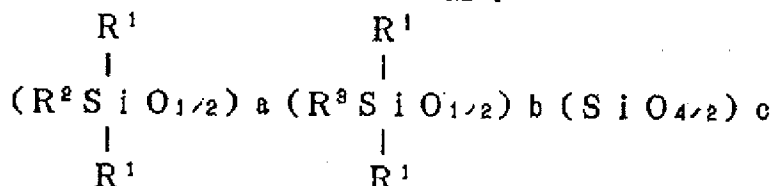
【0005】しかし、特開昭56-145942号公報および特開昭56-136816号公報により提案された硬化性樹脂組成物は、二官能性シロキサン単位(D単位)を必須とするオルガノポリシロキサンを使用しているので、得られた硬化樹脂のガラス転移点(Tg)が低下するという欠点があり、特に、エポキシ基を有する $(CH_3)_2SiO_{2/2}$ 単位(D単位)からなるオルガノポリシロキサンをロキサンを使用する場合は顕著であった。特開昭60-58425号公報により提案された硬化性樹脂組成物は、分子鎖側鎖にエポキシ基とポリオキシアルキレン基を有するジメチルポリシロキサンを使用

するため、得られた硬化樹脂は吸水性が大きく、電気・電子素子用封止樹脂としては適さないという欠点があった。さらに、特開昭64-51467号公報により提案された硬化性樹脂組成物は、これを硬化して得られた硬化樹脂の可撓性が優れるものの、硬化性樹脂組成物の流動性が悪いため、電気・電子素子を樹脂封止する作業性が悪く、さらにこの硬化樹脂は電気・電子素子に対する接着性が悪いという欠点があった。

【0006】

本発明は、(A)硬化性樹脂

および、(B)一般式：



〔式中、 R^1 はアルケニル基を除く一価炭化水素基であり、 R^2 はアルケニル基を除く一価炭化水素基または水素原子であり、 R^3 はエポキシ基含有有機基またはアルコキシシリルアルキル基であり、また a は0または正数であり、 b は正数であり、 c は正数であり、 a/c は0～4の数であり、 b/c は0.05～4の数であり、かつ $(a+b)/c$ は0.2～4の数である。〕で表される、エポキシ基含有有機基を有するオルガノポリシロキサン

0.1～500重量部

からなる硬化性樹脂組成物に関する。

【0009】以下、本発明の硬化性樹脂組成物について詳細に説明する。

【0010】(A)成分は本発明の基材である、硬化性樹脂である。(A)成分としては、従来公知の硬化性樹脂を使用することができる。(A)成分として、具体的には、フェノール樹脂、ホルムアルデヒド樹脂、キシレン樹脂、キシレン-ホルムアルデヒド樹脂、ケトン-ホルムアルデヒド樹脂、フラン樹脂、尿素樹脂、イミド樹脂、メラミン樹脂、アルキッド樹脂、不飽和ポリエステル樹脂、アニリン樹脂、スルホン-アミド樹脂、シリコーン樹脂、エポキシ樹脂およびこれらの共重合体樹脂が例示され、上記例示の硬化性樹脂の1種もしくは2種以上を組み合わせ使用することができる。特に、(A)成分としては、エポキシ樹脂、フェノール樹脂、イミド樹脂およびシリコーン樹脂からなる群から選択される硬化性樹脂であることが好ましい。なお、(A)成分は、その硬化手段が特に限定されず、熱により硬化する硬化性樹脂、紫外線または放射線等の高エネルギー線により硬化する硬化性樹脂、湿気により硬化する硬化性樹脂等が例示される。また、(A)成分は室温で液状または固体状のいずれの状態の硬化性樹脂であってもよい。

*【発明が解決しようとする問題点】本発明者らは、上記問題点を解決するため鋭意努力した結果、本発明に到達した。

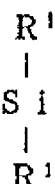
【0007】すなわち、本発明の目的は、硬化前は流動性が優れ、硬化後は可撓性および接着性が優れた硬化樹脂となる硬化性樹脂組成物を提供することにある。

【0008】

【問題点を解決するための手段およびその作用】

100重量部

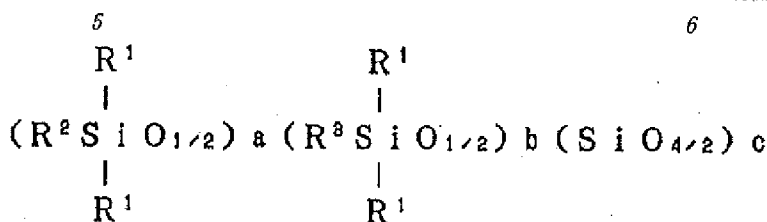
※ ※【化2】



【0011】(A)成分には、上記例示の硬化性樹脂以外の成分として、硬化剤、硬化促進剤、充填剤、光増感剤、高級脂肪酸金属塩、エステル系ワックス、可塑剤等を配合することができる。硬化剤としては、具体的には、カルボン酸やスルホン酸等の有機酸およびその無水物、有機ヒドロキシ化合物、シラノール基、アルコキシ基またはハロゲン基等の基を有する有機ケイ素化合物、一級または二級のアミノ化合物を例示することができ、これらの一種または二種以上を組み合わせ使用することができる。また、硬化促進剤としては、具体的には、三級アミン化合物、アルミニウムやジルコニウム等の有機金属化合物、ホスフィン等の有機りん化合物、異環型アミン化合物、ホウ素錯化合物、有機アンモニウム塩、有機スルホニウム塩、有機過酸化物等が例示される。また、充填剤としては、具体的には、ガラス繊維、石棉、アルミナ繊維、アルミナとシリカを成分とするセラミック繊維、ボロン繊維、ジルコニア繊維、炭化ケイ素繊維、金属繊維、ポリエステル繊維、アラミド繊維、ナイロン繊維、フェノール繊維、天然の動植物繊維等の繊維状充填剤、熔融シリカ、沈澱シリカ、ヒュームドシリカ、焼成シリカ、酸化亜鉛、焼成クレイ、カーボンブラック、ガラスビーズ、アルミナ、タルク、炭酸カルシウム、クレイ、水酸化アルミニウム、硫酸バリウム、二酸化チタン、窒化アルミニウム、炭化ケイ素、酸化マグネシウム、酸化ベリリウム、カオリン、雲母、ジルコニア等の粉粒体状充填剤およびこれらの混合物が例示される。

【0012】(B)成分は、本発明の硬化性樹脂組成物を硬化して得られた硬化樹脂の可撓性を向上させ、かつ電気・電子素子に対する接着性を向上させるための成分であり、一般式：

【化3】



で表されるオルガノポリシロキサンである。上式中、 R^1 はアルケニル基を除く一価炭化水素基であり、具体的には、メチル基、エチル基、プロピル基、ブチル基、ペンチル基、ヘキシル基、オクチル基等のアルキル基；フェニル基、トリル基、キシリル基等のアリール基；ベンジル基、フェネチル基等のアラールキル基；クロロメチル基、3, 3, 3-トリフルオロプロピル基等の置換アルキル基等が例示される。 R^2 はアルケニル基を除く一価炭化水素基または水素原子であり、アルケニル基を除く一価炭化水素基として、具体的には、メチル基、エチル基、プロピル基、ブチル基、ペンチル基、ヘキシル基、オクチル基等のアルキル基；フェニル基、トリル基、キシリル基等のアリール基；ベンジル基、フェネチル基等のアラールキル基；クロロメチル基、3, 3, 3-トリフルオロプロピル基等の置換アルキル基等が例示される。 R^3 はエポキシ基含有有機基またはアルコキシシリルアルキル基であり、エポキシ基含有有機基として、具体的には、グリシドキシエチル基、グリシドキシプロピル基、グリシドキシブチル基、3, 4-エポキシシクロヘキシルエチル基、3, 4-エポキシシクロヘキシルプロピル基、3, 4-エポキシノルボルネニルエチル基、2-(3, 4-エポキシ-3-メチルシクロヘキシル)-2-メチルエチル基等が例示され、またアルコキシシリルアルキル基として、具体的には、トリメトキシシリルエチル基、トリエトキシシリルエチル基、メチルジメトキシシリルエチル基、トリメトキシシリルプロピル基、トリエトキシシリルプロピル基、トリメトキシシリルブチル基、メチルジメトキシシリルプロピル基等が例示される。

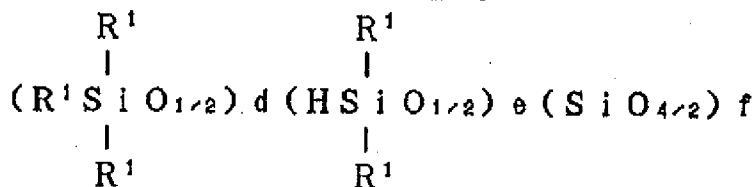
【0013】また、上式中、 a はエポキシ基含有有機基またはアルコキシシリルアルキル基を有しない一官能性*

*シロキサン単位 (M単位) の数を示す0または正数であり、 b はエポキシ基含有有機基またはアルコキシシリルアルキル基を有する一官能性シロキサン単位 (M単位) の数を示す正数であり、 c は四官能性シロキサン単位 (Q単位) の数を示す正数であり、それぞれの比、 a/c は0~4の範囲の数であり、 b/c は0.05~4の範囲の数であり、かつ $(a+b)/c$ は0.2~4の範囲の数である。これは、四官能性シロキサン単位 (Q単位) 1個に対して、一官能性シロキサン単位 (M単位) は4個をこえることはできず、また本発明において、(B)成分が(A)成分に対して相溶性が優れるために、エポキシ基含有有機基またはアルコキシシリルアルキル基を有する一官能性シロキサン単位 (M単位) は、四官能性シロキサン単位 (Q単位) 1個に対して少なくとも0.05個であることが必要であるからである。(B)成分のオルガノポリシロキサンは室温で液状または固体状であり、その分子量は特に限定されないが、(A)成分との相溶性が良好であることから、500~500,000の範囲であることが好ましい。

【0014】本発明において、(B)成分のオルガノポリシロキサンは、エポキシ基含有有機基またはアルコキシシリルアルキル基を有するので、(A)成分の樹脂マトリックス中に取り込まれることにより、得られた硬化樹脂の可撓性が向上し、また(B)成分が SiO_2 単位 (Q単位) からなるので、得られた硬化樹脂のガラス転移点 (T_g) は悪化せず、耐熱性および難燃性に優れるという利点を有する。

【0015】上記(B)成分のオルガノポリシロキサンは、例えば、ヒドロシリル化反応用触媒の存在下、一般式：

【化4】



{式中、 R^1 はアルケニル基を除く一価炭化水素基であり、 d は0または正数であり、 e は正数であり、 f は正数であり、 d/f は0~4の数であり、 e/f は0.05~4の数であり、かつ $(d+e)/f$ は0.2~4の数である。}で表される、ケイ素原子結合水素原子を有するオルガノポリシロキサンとエポキシ基および脂肪族不飽和結合を有する有機化合物と任意量のアルコキシシ

リルアルケンを付加反応させることにより得ることができる。

【0016】(B)成分の配合量は、(A)成分100重量部に対して0.1~500重量部であり、好ましくは0.5~100重量部である。これは、(B)成分の配合量が、(A)成分100重量部に対して0.1重量部未満であると、硬化樹脂の低応力化に効果がないためであり、

また500重量部を超えると硬化樹脂の機械的強度が著しく低下するためである。

【0017】本発明の硬化性樹脂組成物は、(A)成分と(B)成分を均一に混合することにより得られる。(A)成分と(B)成分を混合する方法は特に限定されず、具体的には、(A)成分に直接(B)成分を配合する方法、(A)成分調製時に(B)成分を配合し、プレミックスした後、(A)成分に充填剤等の添加剤を配合する方法、(B)成分に(A)成分に配合する各種の添加剤を順次配合する方法が例示される。(A)成分と(B)成分を混合する混合装置としては、特に限定がなく、(A)成分または(B)成分が液状、固体状、粉状等の種々の形態により混合装置は選択されるが、具体的には、一軸または二軸の連続混合機、二本ロール、ロスミキサー、ニーダーミキサーが例示される。

【0018】本発明の硬化性樹脂組成物は、硬化前には流動性に優れるので、トランスファーモールド、インジェクションモールド、ポッティング、キャストイング、粉体塗装、浸漬塗布、滴下等の方法により使用することができる。また、本発明の硬化性樹脂組成物は、硬化後は可撓性および接着性に優れた硬化樹脂になるので、電気・電子素子を封止するための硬化性樹脂組成物、接着剤等に使用することができる。

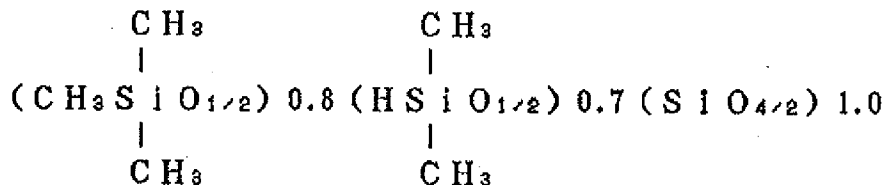
【0019】

【実施例】以下、本発明を実施例により説明する。なお、実施例中、粘度の値は25℃において測定した値であり、硬化性樹脂組成物および硬化樹脂の諸特性は以下に示す方法により測定した。

【0020】○スパイラルフロー：EMMI規格に準じた方法により測定した。

○成形収縮率：JIS-K-6911に準じた方法により測定した。

○熱膨張率：5mm×5mm×16mmに成形した硬化樹脂を熱膨張計（真空理工社製DL-7000）を使*



で表されるオルガノポリシロキサン（粘度105センチポイズ、ケイ素原子結合水素原子の含有量0.40重量%）をトルエン中で、アリルグリシジルエーテル（オルガノポリシロキサン中のケイ素原子結合水素原子のモル数に対して、アリルグリシジルエーテル中のアリル基のモル数は過剰量である。）を塩化白金酸触媒の存在下で反応させて、褐色透明なオルガノポリシロキサンを調製

*用して測定した。熱膨張率の値は、室温からガラス転移点までの値である。

○ガラス転移点(Tg)：熱膨張率測定により測定した。

○曲げ弾性率：JIS-K-6911に準じた方法により測定した。

○吸水率：2インチ×0.5インチ×0.25インチに成形した硬化樹脂を温度121℃、湿度100%にして20時間加湿した後、その硬化樹脂の重量増加を測定することにより求めた。

○バリ：深さ20μmの溝を使用して、バリ長さを測定した。

○接着性：硬化性樹脂組成物を42アロイ板および銅板に挟み、これを硬化させた後、硬化樹脂を剥す際の接着性を○（良好）、×（不良）により判断した。

○耐熱衝撃性：チップサイズ36mm²、パッケージ厚さ2.0mmの樹脂封止半導体素子20個を成形し、これを-196℃→+150℃で1分間のサイクルでヒートサイクル試験を行い、150サイクル後に封止樹脂表面を実体顕微鏡で観察して、表面にクラックの入った成形品の個数が5個以下である場合を○、6~10個である場合を△、11個以上である場合を×とした。

○半田耐熱性：チップサイズ36mm²、パッケージ厚さ2.0mmの樹脂封止半導体素子20個を成形し、これを85℃で85%RH条件下にて72時間放置後、直ちに240℃の半田浴に1時間浸漬し、封止樹脂表面を実体顕微鏡で観察した。表面にクラックの入った成形品の個数が5個以下である場合を○、6~10個である場合を△、11個以上である場合を×とした。

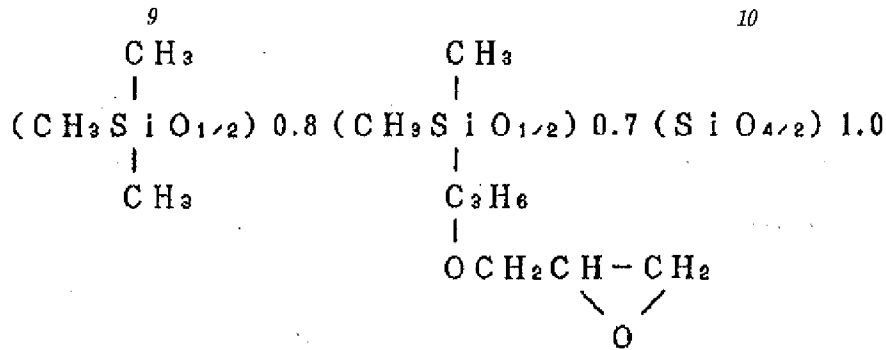
【0021】

【参考例1】攪拌装置、還流冷却管および温度計付きの四つ口フラスコに、式：

【化5】

した。このオルガノポリシロキサンの粘度は520センチポイズであり、エポキシ当量は420であった。このオルガノポリシロキサンは、¹H-核磁気共鳴スペクトル、¹³C-核磁気共鳴スペクトルおよび²⁹Si-核磁気共鳴スペクトルにより、下式で表されるオルガノポリシロキサンであることが確認された。

【化6】

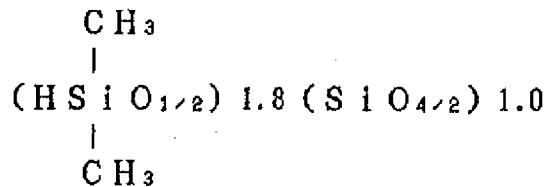


[0022]

*四つ口フラスコに、式：

【参考例2】攪拌装置、還流冷却管および温度計付きの*

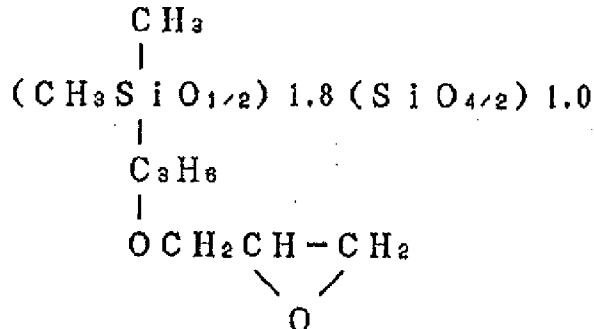
【化7】



で表されるオルガノポリシロキサン（粘度46センチポイズ、ケイ素原子結合水素原子の含有量0.92重量%）をトルエン中で、アリルグリシジルエーテル（オルガノポリシロキサン中のケイ素原子結合水素原子のモル数に対してアリルグリシジルエーテル中のアリル基のモル数が過剰量である。）を塩化白金酸触媒の存在下で反応させて、褐色透明なオルガノポリシロキサンを調製し※

※た。このオルガノポリシロキサンの粘度は610センチポイズであり、エポキシ当量は370であった。このオルガノポリシロキサンは、 ^1H -核磁気共鳴スペクトル、 ^{13}C -核磁気共鳴スペクトルおよび ^{29}Si -核磁気共鳴スペクトルにより、下式で表されるオルガノポリシロキサンであることが確認された。

【化8】

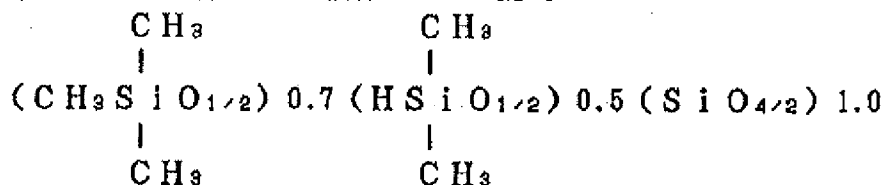


[0023]

★四つ口フラスコに、式：

【参考例3】攪拌装置、還流冷却管および温度計付きの★

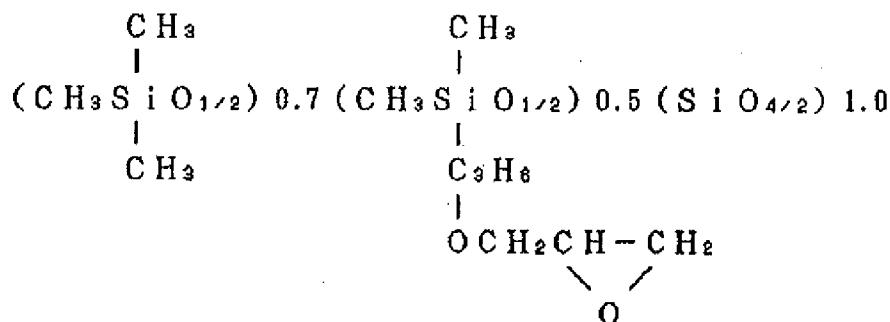
【化9】



で表される粘稠なオルガノポリシロキサン（ケイ素原子結合水素原子の含有量0.33重量%）をトルエン中で、アリルグリシジルエーテル（オルガノポリシロキサン中のケイ素原子結合水素原子のモル数に対してアリルグリシジルエーテル中のアリル基のモル数が過剰量である。）を塩化白金酸触媒の存在下で反応させて、褐色半

透明な水飴状のオルガノポリシロキサンを調製した。このオルガノポリシロキサンのエポキシ当量は1100であった。このオルガノポリシロキサンは、 ^1H -核磁気共鳴スペクトル、 ^{13}C -核磁気共鳴スペクトルおよび ^{29}Si -核磁気共鳴スペクトルにより、下式で表されるオルガノポリシロキサンであることが確認された。

【化10】

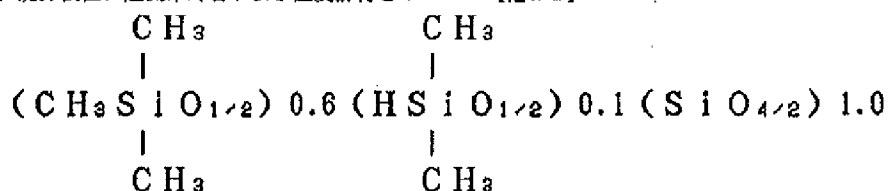


【0024】

*四つ口フラスコに、式：

【参考例4】攪拌装置、還流冷却管および温度計付きの*

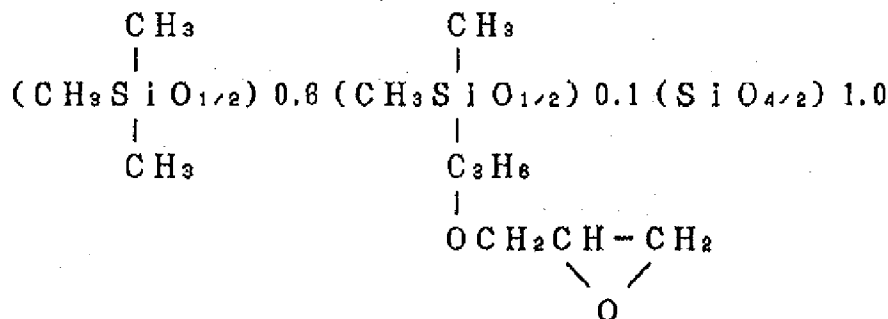
【化11】



で表されるオルガノポリシロキサン（ケイ素原子結合水素原子の含有量0.09重量%）の68重量%のトルエン溶液に、アリルグリシジルエーテル（オルガノポリシロキサン中のケイ素原子結合水素原子のモル数に対してアリルグリシジルエーテル中のアリル基のモル数が過剰量である。）を塩化白金酸触媒の存在下で反応させて、褐色半透明な水飴状のオルガノポリシロキサンを調製し※

20※た。このオルガノポリシロキサンのエポキシ当量は1290であった。このオルガノポリシロキサンは、 ^1H -核磁気共鳴スペクトル、 ^{13}C -核磁気共鳴スペクトルおよび ^{29}Si -核磁気共鳴スペクトルにより、下式で表されるオルガノポリシロキサンであることが確認された。

【化12】

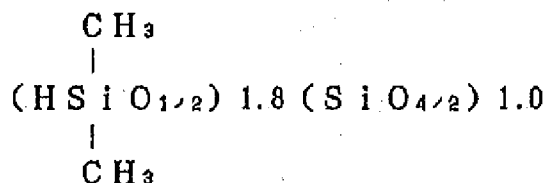


【0025】

★四つ口フラスコに、式：

【参考例5】攪拌装置、還流冷却管および温度計付きの★

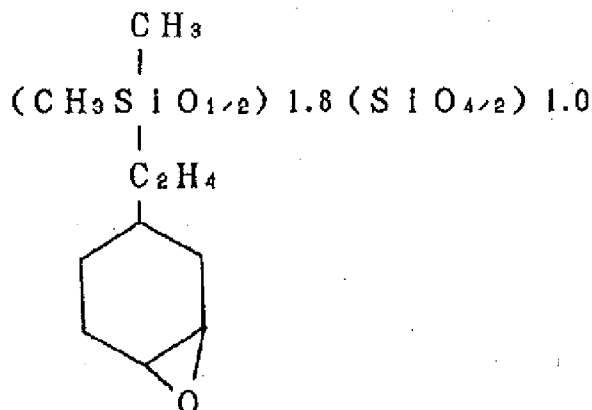
【化13】



で表されるオルガノポリシロキサン（粘度46センチポイズ、ケイ素原子結合水素原子の含有量0.92重量%）をトルエン中で、1,2-エポキシ-4-ビニルシクロヘキサン（オルガノポリシロキサン中のケイ素原子結合水素原子のモル数に対して、1,2-エポキシ-4-ビニルシクロヘキサン中のビニル基のモル数が過剰量

である。）を塩化白金酸触媒の存在下で反応させて、褐色透明のオルガノポリシロキサンを調製した。このオルガノポリシロキサンの粘度は520センチポイズであり、エポキシ当量は230であった。このオルガノポリシロキサンは、 ^1H -核磁気共鳴スペクトル、 ^{13}C -核磁気共鳴スペクトルおよび ^{29}Si -核磁気共鳴スペクトル

により、下式で表されるオルガノポリシロキサンである * 【化14】
ことが確認された。 *

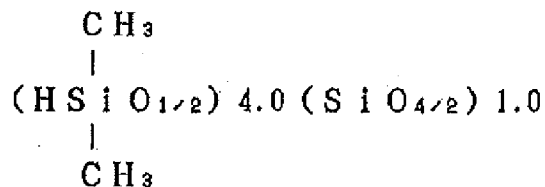


【0026】

※四つ口フラスコに、式：

【参考例6】攪拌装置、還流冷却管および温度計付きの※

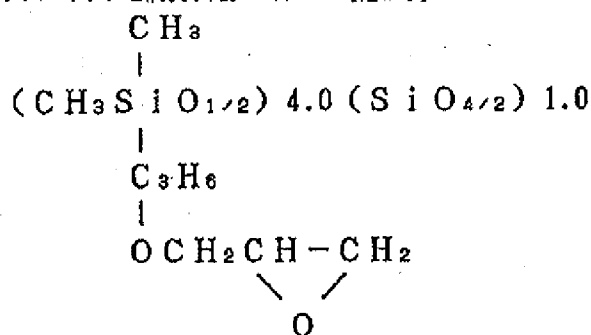
【化15】



で表されるオルガノポリシロキサン（沸点190℃、ケイ素原子結合水素原子の含有量1.22重量%）をトルエン中で、アリルグリシジルエーテル（オルガノポリシロキサン中のケイ素原子結合水素原子のモル数に対して、アリルグリシジルエーテル中のアリル基のモル数が過剰量である。）を塩化白金酸触媒の存在下で反応させて、褐色透明のオルガノポリシロキサンを調製した。こ★

★のオルガノポリシロキサンの粘度は54センチボイズであり、エポキシ当量は205であった。このオルガノポリシロキサンは、 ^1H -核磁気共鳴スペクトル、 ^{13}C -核磁気共鳴スペクトルおよび ^{29}Si -核磁気共鳴スペクトルにより、下式で表されるオルガノポリシロキサンであることが確認された。

【化16】

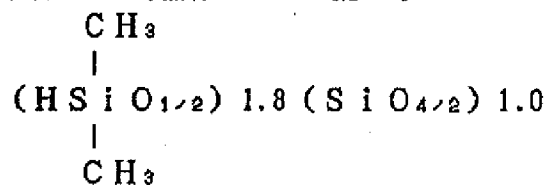


【0027】

☆四つ口フラスコに、式：

【参考例7】攪拌装置、還流冷却管および温度計付きの☆

【化17】



で表されるオルガノポリシロキサン（粘度46センチボイズ、ケイ素原子結合水素原子の含有量0.92重量%）をトルエン中で、アリルグリシジルエーテルと同モルのアリルトリメトキシシラン（オルガノポリシロキサ

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* Nは、 ^1H -核磁気共鳴スペクトル、 ^{13}C -核磁気共鳴スペクトルおよび ^{29}Si -核磁気共鳴スペクトルにより、下式で表されるオルガノポリシロキサンであることが確認された。

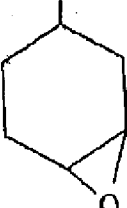
$$\begin{array}{ccc} \text{CH}_3 & & \text{CH}_3 \\ | & & | \\ (\text{CH}_3\text{SiO}_{1/2})_{0.9} & & (\text{CH}_3\text{SiO}_{1/2})_{0.9}(\text{SiO}_{4/2})_{1.0} \\ | & & | \\ \text{C}_3\text{H}_8 & & \text{C}_3\text{H}_8 \\ | & & | \\ \text{Si}(\text{OCH}_3)_3 & & \text{OCH}_2\text{CH}-\text{CH}_2 \\ & & \diagdown \quad \diagup \\ & & \text{O} \end{array}$$

※ 1 に示した。

[0029]

【化19】

$$\begin{array}{ccccccc} \text{CH}_3 & & \text{CH}_3 & & \text{CH}_3 & & \text{CH}_3 \\ | & & | & & | & & | \\ \text{CH}_3\text{SiO} & (\text{SiO})_{98} & (\text{SiO})_5 & \text{Si} & \text{CH}_3 \\ | & & | & & | \\ \text{CH}_3 & & \text{CH}_3 & & \text{C}_2\text{H}_4 & & \text{CH}_3 \end{array}$$



と同様にして硬化させた。得られた硬化樹脂の諸特性を表1に示した。

[0031]

【表 1】

測定項目	本発明	比較例	
	実施例1	比較例1	比較例2
スパイラルフロー (インチ)	36	35	31
成形収縮率 (%)	0.22	0.20	0.31
熱膨張率 ($\times 10^{-5}/^{\circ}\text{C}$)	1.10	1.05	1.50
ガラス転移点 ($^{\circ}\text{C}$)	155	135	156
曲げ弾性率 (kg/mm^2)	1210	1250	1350
吸水率 (%)	0.42	0.55	0.51

【0032】

【実施例2】 $\text{CH}_3\text{SiO}_{3/2}$ 単位40モル%、 C_6H_5 5 (CH_3) $\text{SiO}_{2/2}$ 単位10モル%、 $\text{C}_6\text{H}_5\text{SiO}_{3/2}$ 単位40モル%および $(\text{C}_6\text{H}_5)_2\text{SiO}_{2/2}$ 単位10モル%からなり、Siに直結する水酸基を5重量%含有するメチルフェニルポリシロキサン樹脂13重量部とクレゾールノボラックエポキシ樹脂 (軟化点80℃、エポキシ当量220) 13重量部からなるシリコーンエポキシ樹脂に参考例2で調製したオルガノポリシロキサン2重量部、溶融石英粉末74重量部、アルミニウムアセチルアセトネート0.90重量部およびカルナウバワックス1重量部とを90℃の加熱ロールで混練して、硬化性樹脂組成物を得た。次いで、これを粉砕し、175℃、70 kg/cm^2 の条件下で2分間トランスファモールド成形した。その後、この硬化樹脂を180℃で12時間かけてポストキュアした。得られた硬化樹脂の諸特性を表2に示した。

【0033】

【実施例3】 実施例2において、参考例2で調製したオルガノポリシロキサンの代わりに参考例3で調製したオ

ルガノポリシロキサンを使用した以外は実施例2と同様にして、硬化性樹脂組成物を調製した。これを実施例2と同様にして硬化させた。得られた硬化樹脂の諸特性を表2に示した。

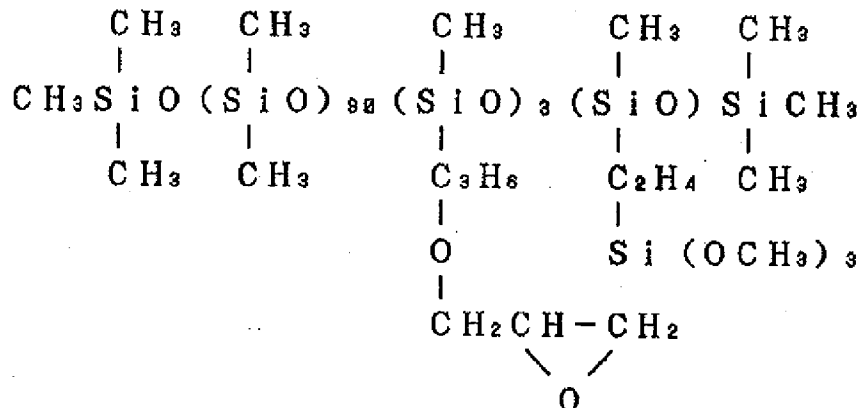
【0034】

【実施例4】 実施例2において、参考例2で調製したオルガノポリシロキサンの代わりに参考例4で調製したオルガノポリシロキサンを使用した以外は実施例2と同様にして、硬化性樹脂組成物を調製した。これを実施例2と同様にして硬化させた。得られた硬化樹脂の諸特性を表2に示した。

【0035】

【比較例3】 実施例2において、参考例2で調製したオルガノポリシロキサンの代わりに下式で表されるオルガノポリシロキサンを使用した以外は実施例2と同様にして硬化させた。得られた硬化樹脂の諸特性を表2に示した。

【化20】



【0036】

【比較例4】実施例2において、参考例2で調製したオルガノポリシロキサンを添加しない以外は実施例2と同様にして硬化性樹脂組成物を調製した。これを実施例2*

*と同様にして硬化させた。得られた硬化樹脂の諸特性を表2に示した。

【0037】

【表2】

測定項目	本 発 明			比 較 例	
	実施例2	実施例3	実施例4	比較例3	比較例4
スパイラルフロー(インチ)	46	43	41	40	35
成形収縮率 (%)	0.34	0.36	0.32	0.37	0.43
熱膨張率 ($\times 10^{-5}/^{\circ}\text{C}$)	2.32	2.25	2.20	2.90	2.75
ガラス転移点 ($^{\circ}\text{C}$)	175	173	176	151	170
曲げ弾性率 (kg/mm^2)	1100	1120	1140	1250	1520
吸水率 (%)	0.43	0.41	0.40	0.56	0.47
バリ (mm)	4	5	3	15	4
接着性	○	○	○	△	×
耐熱衝撃性	○	○	○	△	×
半田耐熱性	○	○	○	×	×

【0038】

【実施例5】ビスマレイミド-トリアジン型の熱硬化性ポリイミド樹脂35重量部、参考例5で調製したオルガノポリシロキサン4重量部、溶融石英粉末65重量部、カルナウバワックス1重量部および安息香酸アルミニウム0.32重量部とを90℃の加熱ロールで混練し、硬化性樹脂組成物を調製した。次いで、これを粉碎して、220℃、70kg/cm²の条件下で4分間トランスファモールド成形した。その後、この硬化樹脂を230℃で3時間かけてポストキュアした。得られた硬化樹脂の諸特

性を表3に示した。

【0039】

【比較例5】実施例5において、参考例5で調製したオルガノポリシロキサンを使用しない以外は実施例5と同様にして硬化性樹脂組成物を調製した。これを実施例5と同様に硬化させた。この硬化樹脂の諸特性を表3に示した。

【0040】

【表3】

測定項目	本発明	比較例
	実施例5	比較例5
スパイラルフロー (インチ)	52	44
曲げ弾性率 (kg/mm ²)	980	1220
接着性	○	×

【0041】

【実施例6】オルソクレゾールノボラックエポキシ樹脂（軟化点80℃、エポキシ当量220）75重量部、溶融シリカ260重量部、カルナバウワックス1重量部、フェノールノボラック樹脂35重量部、トリフェニルフォスフィン0.6重量部、参考例6で調製したオルガノポリシロキサン5重量部を、90℃の加熱ロールで混練して、硬化性樹脂組成物を調製した。次いで、これを粉

*ルガノポリシロキサンに参考例7で調製したオルガノポリシロキサンを使用した以外は実施例6と同様にして硬化性樹脂組成物を調製した。これを実施例6と同様にして硬化させた。得られた硬化樹脂の諸特性を表4に示した。

【0043】

【比較例6】実施例6において、参考例6で調製したオルガノポリシロキサンを使用しない以外は実施例6と同様にして硬化性樹脂組成物を調製した。これを実施例6と同様にして硬化させた。得られた硬化樹脂の諸特性を表4に示した。

【0044】

【表4】

【0042】

【実施例7】実施例6において、参考例6で調製したオ*

測定項目	本発明		比較例
	実施例6	実施例7	比較例6
スパイラルフロー (インチ)	73	70	56
熱膨張率 ($\times 10^{-5}/^{\circ}\text{C}$)	2.70	2.65	2.67
ガラス転移点 ($^{\circ}\text{C}$)	138	140	135
曲げ弾性率 (kg/mm ²)	1150	1210	1420
吸水率 (%)	0.49	0.52	0.53
接着性	○	△~○	×
耐熱衝撃性	○	○	×
半田耐熱性	○	○	×

【0045】

50 【発明の効果】本発明の硬化性樹脂組成物は、(B)成分

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としてエポキシ基結合有機基を有するオルガノポリシロキサンを配合しているので、硬化前は流動性に優れ、こ

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れを硬化して得られた硬化樹脂が可撓性および接着性に優れるという特徴を有する。